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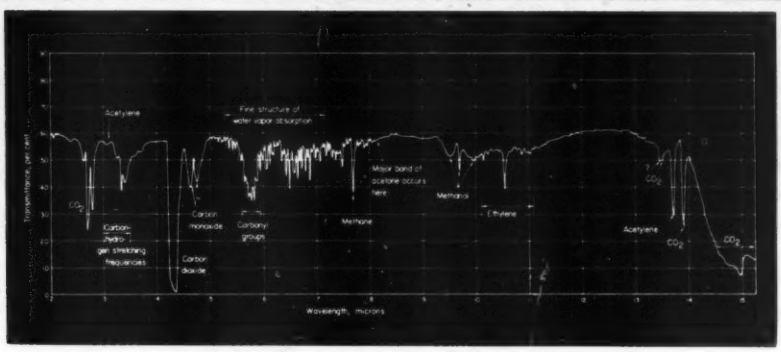

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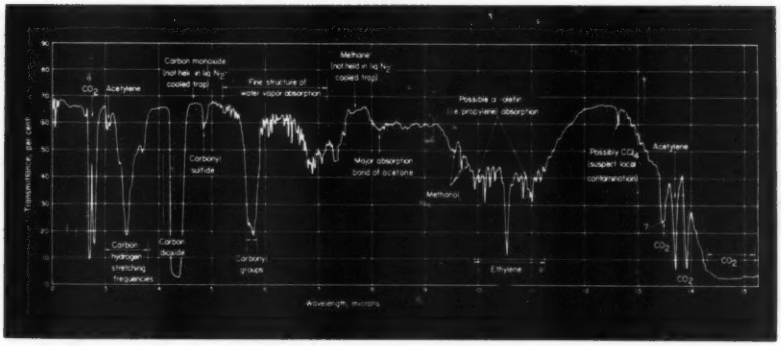

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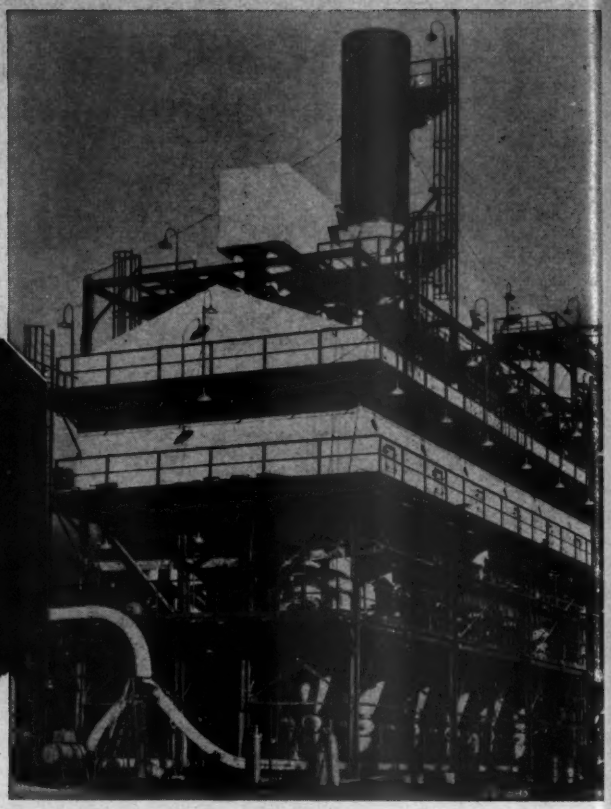

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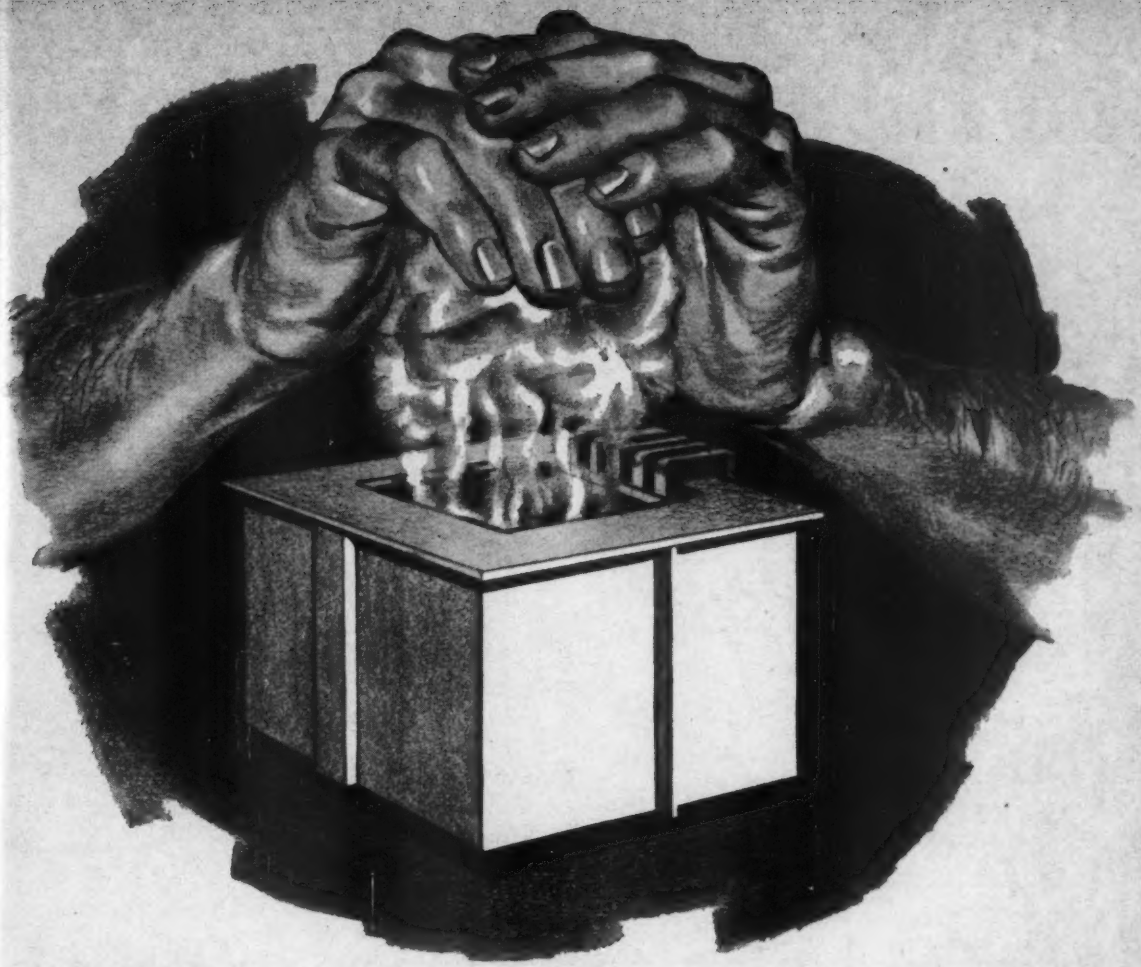

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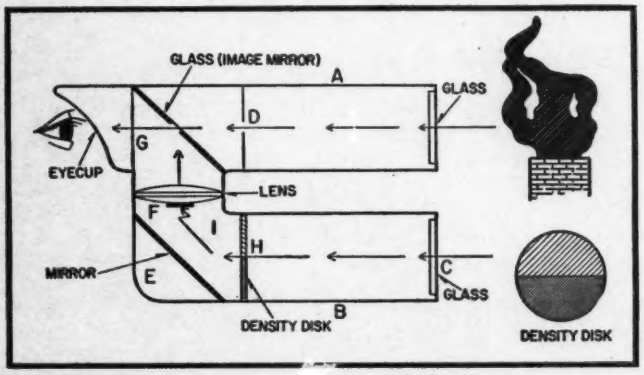

● ***How it works***—Smoke picture fills field of vision through apertures C, D, and G. Light from area adjacent to stack is transmitted through reference disc, H, in barrel, B, to surface of mirror, E. Image of reference disc projects through lens, F, to image mirror where it is compared with smoke seen through apertures. Center of reference disc is blocked by opaque disc, I.

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The Editor welcomes writers interested in air pollution problems and encourages them to use the pages of this journal for the expression of their opinions. But neither he nor the Association assumes responsibility for the validity of the opinions expressed herein.

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Test of "Fiberfrax" Ceramic Fibers for High Temperature Gas Filtration*†

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This study was initiated to develop a commercially available system for removing solid radioactive components from hot exhaust gases discharged from a testing facility. Removal of particulate material from very hot gases (i.e., >1000°F) has considerable importance for the recovery of valuable products (as in the metallurgical industries) and for the economic control of air pollution.

Development of *Fiberfrax*, a fibrous material capable of withstanding temperatures up to 2000°F for prolonged periods, presented an opportunity for investigating some of the practical and theoretical factors involved in the filtration of small particulate materials from gases at temperatures above 1000°F. *Fiberfrax* ceramic fiber, Fig. 1, is a white fibrous material similar in appearance to spun glass. Chemically, it consists of approximately equal parts of alumina and silica plus small percentages of boric oxide, zirconia or other fluxes. It is produced by processes similar to those used for mineral wool or staple glass fibers, except that its melting point (3300°) is so high that the heat of an electric arc furnace is required. The molten mix is formed into fibers by blowing with steam or air, or by spinning from mechanical rotors. The loose masses of fiber can be cleaned and converted into various forms such as paper, insulating blocks, blankets and woven fabrics. Resistence to high temperatures is unexcelled by any fiber with the possible exception of pure silica. It can withstand short exposure to temperatures of 2300°F and prolonged exposure to at least 2000°F. It shows no tendency to melt or sinter even at 2500°F, but gradually changes from an amorphous glassy structure to crystalline during exposure to temperatures over 2000°F. This is a time-temperature effect, proceeding faster at higher temperatures. The effect of devitrification, or crystallization, is to make the fibers more brittle, but in the temperature range below 1500°F fibers are substantially unaffected even by very long exposure.

Fiberfrax ceramic fiber is slightly more resistant to corrosion by acids and alkalies than ordinary glass fibers.

Fig. 1. Filter before use

Strength and resistance to vibration are the same or perhaps slightly less than those of glass fibers. The presence of shot in the material as formed often requires a cleaning operation for removal.

Effect of Elevated Temperatures on Filter Performance

For viscous flow conditions the pressure gradient (P/L) through fiber beds of high porosity takes the following general form, adapted from Sullivan[12]:

$$\frac{P}{L} = \frac{KV\mu}{gd^2\rho} \times \frac{(1-\varepsilon)^2}{\varepsilon^3}$$

where the ratio $(1-\varepsilon)^2/\varepsilon^3$ is the Kozeny voids-correction factor.

P = pressure loss, ft. of fluid flowing
L = bed depth, ft.
V = superficial filtration velocity, ft./sec.
d = fiber diam., ft.
K = a constant whose value is dependent on fiber orientation
g = gravitational constant, ft./sec.^{2}
μ = air viscosity, lb./sec. ft.
= density, lb./ft.^{3}
ε = fraction voids

Considering only the effects of temperature on a given filter the value of K, L, g, d, V and the porosity factor are unaffected and resistance is proportional to the ratio μ/P, (kinematic viscosity); which varies with temperature in accordance with the relationship shown in Fig. 2, Curve A. For equal gas velocities, filter resistance may be expected to rise with increase in temperature of the gases. In the units commonly used to express filter resistance (height of equivalent column of water) the gas density term, ρ, factors out and resistance is proportional to absolute viscosity, μ. On the basis of equal weights of gas, velocity is proportional to absolute temperature and resistance will be proportional to the product of superficial gas velocity and kinematic viscosity. The change in value of the product V (μ/P)

† Reprinted from *Industrial and Engineering Chemistry* 48, 696-702 (April 1956).

* Presented at the 48th Annual Meeting of the Air Pollution Control Association held at Detroit, Mich., May, 1955.

[12] Sullivan, R. R., *J. App. Phys.*, 12, 503 (1941).

with rise in temperature (vol. of gas at 70°F = unity) is shown in Fig. 2, Curve B. It may be noted that filter resistance increases at an especially rapid rate for temperatures above 500°F. Flow conditions in all experiments conducted during this study were within the viscous range for which Equation 1 was believed to be accurate.

Collection of Particles

Collection of particles is accomplished by several mechanisms depending on size and character. Particles greater than about 1 μ in size are captured principally by impaction while smaller particles (i.e., smaller than 0.5 μ) are separated most effectively by diffusional and electrostatic forces. It was anticipated that electrostatic forces were not likely to be an important factor at temperatures above 1000°F and since the test dust used in the experimental study contained a negligible weight fraction below 0.5 μ in size, the principal interest was in the effect of high temperatures on the operation of the inertial mechanism.

Sell and Albrecht (reported by Johnstone and Roberts)[9] have shown impaction efficiency to be a function of the dimensionless group

$$\frac{mv}{kd}$$

For spherical particles: $\frac{mV}{kd} = \frac{1}{18}\frac{D^2V\rho_s}{\mu d}$

Where

m = mass of aerosol particle
D = diameter of aerosol particle
ρs = density of particles
k = Stokes' law resistance coefficient ($k = 3\pi\mu D$ for spheres) and other symbols are as noted above.

Although solid particles are rarely spherical in practice, (the value of k in the above equation may require some modification in these cases) the viscosity factor is likely to remain unchanged so long as particles are moving in the viscous flow range. Therefore impaction efficiency (E) of small fibers for small particles is inversely proportional to some function of the absolute viscosity of the gas (all other factors being constant) and decreases as the temperature of the gas rises. For constant gas mass velocity, impaction efficiency is proportional to some function of the ratio V/μ. The exact relationship between impaction efficiency of a cylindrical body—e.g., a fiber—and the dimensionless group for spherical particles has been determined by a number of investigators.

Using the curves of Langmuir and Blodgett (reproduced by Lapple[10] and reported by him to be reliable) the effect on impaction efficiency of a temperature rise from 70° to 1500°F may be estimated. As the curve has many changes in slope, the effects of a rise in temperature are not uniform over the entire range. For example, for 1 μ spherical particles of specific gravity 6.4, filter fiber diam. of 20 μ, and an air flow rate of 350 ft./min. (typical values for this study), impaction efficiency decreases from 68 to 50% when temperature rises from 70° to 1500°F and gas flow rate remains constant—i.e., 350 ft./min. at both temperatures. If the gas mass rate of flow remains constant, but otherwise conditions are the same as noted above, target efficiency increases from 68% at 70°F to 80% at 1500°F. A similar calculation for particles 10 μ in diam. shows target efficiencies > 99% at both temperatures for constant gas volume and constant gas mass rate conditions.

Experimental Equipment

Special care was required in the selection of materials of construction for the test equipment because tests were to be conducted at temperatures up to 1500°F. Fig. 3 shows a schematic diagram of the assembled apparatus.

Room air was blown through the electric air-heating furnace and to the test duct with a blower. Arrangements were made at the fan inlet to filter the test air stream and to regulate air volume by means of a damper.

The air heater was in the form of a steel-clad, fire-brick-lined box approximately 3 ft. wide, 5 ft. long and 3 ft. high containing 28 1-in. diam. Globar silicon carbide heating elements and 14 vertical baffles so that the air traveled a total linear path of 28 ft. in passing through the heating chamber. With a power input of 70 kw. it was possible to continuously heat 175 cfm. of room air (entering at approximately 70-100°F) to a temperature of 1500°F.

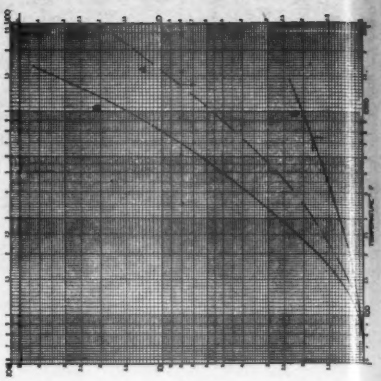

Fig. 2.

Production of high air temperatures by combustion of gases such as oxygen and acetylene in the test duct was considered to be somewhat easier and considerably cheaper, but electric heating was preferred because of ease of temperature control and complete absence of smoke and soot.

The test duct was 6 in. square on the inside and was constructed of 12 gage #310-stainless steel. This material was selected because of its excellent heat resistance. For dimensional stability the 13 ft. long tunnel was reinforced with 2x2x1/4 in. #310-stainless steel angles welded the entire length along 2 sides. The test duct was constructed in 3 sections (7, 4 and 2 ft. long) with provisions for mounting a filter between the 2 largest sections and a flow meter between the 2 smallest pieces. Both ends of each section were flanged with 2x2x1/4 in. angles suitably drilled for bolting. When assembled, a short transition piece connected the furnace and 7 ft. section of duct. At the furnace end there was an opening for introducing the test dust. Three ft. downstream there was a centrally located stop[2] which served to thoroughly mix dust and air and produced a uniform duct velocity and dust concentration profile at the sampling station.

On the downstream end of the 7 ft. duct were pressure and thermocouple taps and an opening for admitting the upstream sampling probe. The upstream end of the 4 ft. section contained a pres-

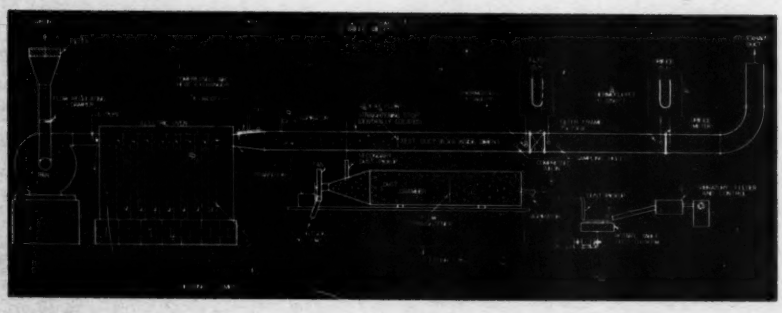

Fig. 3.

[9] Johnstone, H. F. and Roberts, M. H., *Ind. Eng. Chem.* 41, 2417 (1949).

[2] Anonymous. (Stairmand, C. J.) *Eng.* (London) 152, 141 (1941).

Fig. 4.

sure tap and opening for introducing the downstream sampling probe while the opposite end contained thermocouple and orifice meter pressure taps. The smallest section contained only a single orifice meter pressure tap at the upstream end. The entire test duct and filter frame were enclosed in a 3 in. thick insulating blanket composed of *Fiberfrax* brick plus metal foil. Fig. 4 shows the insulated test duct with filter removed. Air heater, flow meters, sampling probes, *etc.*, are also shown in the photo.

Filter frames were constructed of 18 gage #310-stainless steel and were 7 in. square in cross section. There was a ½ in. lip on each end so that the actual opening was 6 x 6 in. (to match the inside dimensions of the test duct). Filter frames were 8 in. deep but provision was made to accommodate filter depths from 1 to 8 in. Stainless steel screening (¼ in. mesh) was used on front and back surfaces of the filter.

Dust feeding mechanism for producing a steady flow of a well dispersed, non-agglomerated dry powder to the test duct consisted of a vibratory feeder which dropped a ribbon of dust onto a rotating turntable. A flow-regulating scraper allowed material to pass at a uniform rate, while the excess dropped to

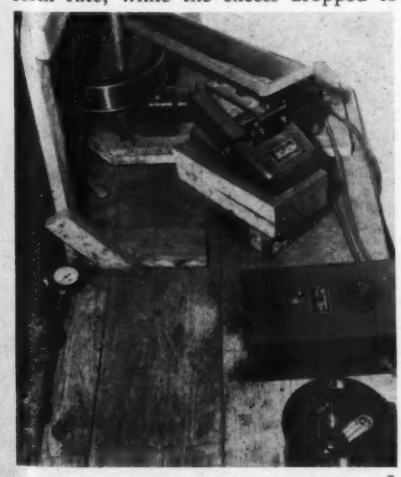

Fig. 5.

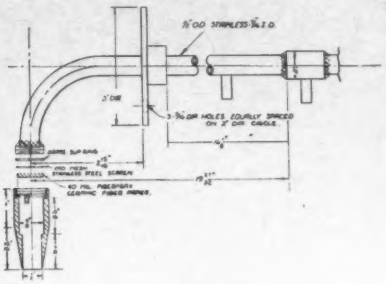

Fig. 6.

a bin below. The uniform ribbon of dust passing the scraper was raised in a pipe by induced air from an aspirator supplied with primary air at a pressure of 10-25 lb. gage. The apparatus is shown in Fig. 5, including vibrator control box. A more detailed description of the apparatus is given in a previous publication.[4]

For filter testing, dust loadings in the order of 1 gr./1000 ft.[3] were desired. Since steady concentrations of this low value were not easily obtained with the above apparatus, a dust dispersion and dilution chamber (a 4 ft. length of 15 in. diameter pipe containing a centrally located baffle about 3 ft. from the entry) was added to further dilute the aerosol and to settle out dust agglomerates which resisted the disruptive action of the first jet. At the extreme downstream end of the dilution chamber a small aliquot of the dispersion was picked up by a second aspirator and blown into the test duct (while the excess was blown to the out-of-doors by a small blower). Dust concentrations in the test duct were controlled by adjusting (1) the amount of dust picked up from the turntable and (2) the size of the fraction removed from the dust chamber.

It was found that air entering the test duct through the final dust aspirator produced a serious cooling effect upon the hot gases leaving the furnace. To remedy this, the compressed air supply (primary jet air) was warmed by passing it through a flat, stainless steel tube-coil placed directly in contact with the hot metal surfaces of the transition piece on the exit side of the furnace. This device may be seen in place in Fig. 4.

Flow metering of air in the test duct was accomplished with the aid of a 1 in. diameter sharp-edged, centrally located orifice bolted between the flanges of 2 sections of test duct and calibrated in

[1] Allen, G. L., Viets, F. H. and McCabe, L. C., U.S. Bur. of Mines Inform. Circ. 7627, April 1952.

[2] See footnote 2, page 62.

[4] First, M. W., Silverman, L., et al. Report No. NYO-1581, Harvard Univ. 1952.

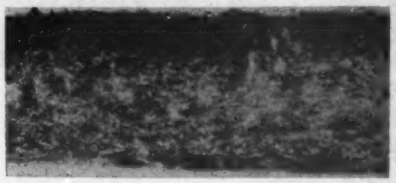

Fig. 7 Probe pad down stream Mag. x 54

place with the aid of a standard Pitot-static tube. Flow was corrected for temperature with the aid of a Chromel-Alumel thermocouple placed just upstream of the orifice plate. A similar thermocouple was placed just ahead of the test filter to measure the temperature of the air at this point also.

Up-and-downstream air samples were withdrawn simultaneously through stainless steel probes shown in Fig. 6. Dust samples were collected isokinetically on filters held in the tip of the probe and faced upstream. To withstand the heat, filters were cut from special *Fiberfrax* paper. This paper was approximately 0.040 in. thick and was composed largely of fibers less than 1 μ in diameter. It has an efficiency of 99.9+% for 0.3 μ DOP smoke.

The paper was somewhat fragile and had to be handled carefully. When placed in the sample probe it was backed with a 200 mesh stainless steel screen for support and enclosed in a stainless steel slip ring to prevent twisting and tearing of the filter when the tip and probe were screwed tightly together to form an air-tight edge seal. The test dust used in the efficiency studies was dark brown in color, consequently any edge leakage or penetration through the filter was immediately evident. In no instance was penetration through or around a sample filter noted. Fig. 7 is a section through one of the sampling filters and shows little dust penetration. Provision was also made to use a plug of random packed fibers in a recess in the head of the probe, but the paper discs proved so much simpler to use that other methods were not utilized to any great extent.

The sample filters placed in the tip of the probe were 11/16 in. in diam. and weighed approximately 60 mg. when new. Filters were weighed cold before and after sampling along with a number of blanks to control small daily variations in the amount of adsorbed moisture. This correction rarely exceeded a few tenths of a milligram. A significant weight loss occurred on exposure to high temperatures, attributed to loss of a small percent of a volatile binder used in manufacturing the paper. Weight loss after exposure to high temperature was found to average 1.6 mg. for filters whose original weight was between 50 and 60 mg. The weight

Fig. 8. Fiberfrax

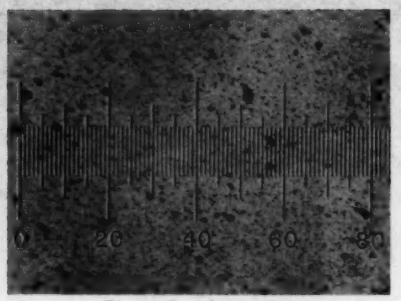

Fig. 9. One division = cupric oxide mag. X100

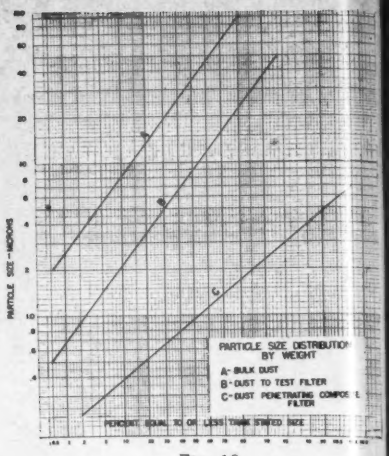

Fig. 10.

increase of the sample filter (after suitable corrections) plus sample flow rate and time of the test permitted calculation of dust loading up and down stream of the test filter and filtration efficiency.

A stainless steel Venturi meter was welded onto each sampling probe about 12 in. back from a 90° elbow and seamless copper tubing, brazed to the downstream end of the Venturi meter, completed the run to a compressed air aspirator. The pressure leads from the Venturi meter were seamless copper tubing brazed to stainless steel meter taps. In order to obtain flexibility required to insert or remove sampling probes, the copper pressure leads and probe extension were provided with gas-tight couplings which could be connected or disconnected with a few turns of a wrench.

Thermocouple taps were provided just upstream of the flow meters so that readings could be corrected for gas temperature. It was noted that in passing through the 12 or so inches of ½ in. diam. seamless stainless steel tubing exposed to the ambient air that the 1400° gases were cooled to less than 150°F. Had this rapid cooling of the sampled gases been anticipated, somewhat simpler arrangements for connecting the probe and meter to a source of suction and a liquid manometer might have been utilized.

Samples for determination of particle size distribution were taken up and down stream of the test filter with the aid of membrane filters[7] and analyzed microscopically. Size analyses of test dusts scraped off the surface of the high temperature sample filters were in fair agreement with the low temperature membrane filter samples. However, the latter were considered more accurate because there was no agglomeration during sampling nor shattering during the preparation of the microscope mount.

Filter resistance of each test filter was noted at a number of air flow rates under hot and cold conditions.

Test filters were prepared from 3 different fiber sizes. Size and size distribution data for each fiber are indicated in Table I and photomicrographs of the fibers are shown in Fig. 8. Weight percentage of shot in each of the experimental fibers was approximately 65%. Considerable improvement in proportion of fibers to shot would be possible in quantity production of similar fibers. It is believed that the presence of shot did not seriously affect the conduct of the tests or the results. Filter packing density was calculated on the basis of the weight of effective fibers rather than total weight of fibers plus shot.

Studies carried out to determine the smallest size test filter which will indicate the performance of full scale filters have shown[5] that as the filter becomes smaller in cross-section (and the perimeter to area ratio larger) the influence of discontinuities in bed porosity becomes very great and turbulent or transition type flow occurs through the edges of the bed. A comparison of resistance measurements of 6 x 6 in. test filters with those 12 x 12 and 20 x 20 in. showed no significant differences, while a marked variation was observed in filters smaller than 6 x 6 in. face area. Therefore, it was concluded that a 6 x 6 in. test chamber would be the minimum size suitable for measuring the resistance of deep bed fiber filters. The filter frames used in this study were 7 x 7 in. in cross-section but had an open face area of only 6 x 6 due to the presence of a half inch lip around the perimeter of each end of the box.

Because of the possibility of a separation between medium and frame, due to differential expansion of the ceramic and metal during the heating process, it was routine practice during this investigation to line the inner surfaces of the filter holder with a thin layer of small diameter fibers to serve as a gasket or seal. This practice produced consistent results during testing and more nearly represented full-scale filter performance wherein minor discontinuities at the filter frame wall become a much smaller factor in overall performance (because of the small size of the perimeter compared to the filter cross-section).

In addition to measuring resistance and dust removal efficiency, studies were conducted to evaluate service life under conditions of vibration and cyclic heating and cooling, and the dust retention ability of dust-loaded fibers was also determined.

The test dust used in all efficiency experiments was Baker's technical grade black copper oxide (CuO) having a true specific gravity of 6.4. This material presented an irregular appearance under the microscope (Fig. 9) and showed a wide range of particle sizes. Fig. 10, Curve A, shows the size distribution of the bulk material. Fig. 10, Curve B, is the size distribution of the dust cloud actually blown into the test duct and indicates that considerable portions of the heavy fractions were removed by impingement on the baffle and by settling in the 15 in. diam. dilution chamber noted in Fig. 4. The dust cloud entering the filter had a mass median diameter of 8.5 μ. Sixty-nine percent by weight of

[7] First, M. W. and Silverman, L. *AMA Arch. Ind. Hyg. & Occ. Med.* 7, 1 (1953).

[5] First, M. W., Silverman, L. et. al. Report No. NYO 1591, Harvard Univ. 1954.

TABLE I
Size of Filter Fibers

Fiber	Mean diam. μ	Maximum diam. μ	Minimum diam μ
Fine	4	40	2
Medium	8	40	4
Coarse	20	80	4

the suspension was greater than 5μ in size and 97.5% was greater than 1 μ, indicating that this was a typical industrial dust in size and within the range of commonly encountered specific gravity.

Conduct of all tests was as follows: A test filter was prepared by weighing out the required quantity of fibers and hand packing the frames with special attention to the corners and edges. Gaskets of *Fiberfrax* paper were cemented with water glass to each face of the frame and then bolted, air tight, between the flanges of the test duct. Next, the main air supply blower was started and the duct and filter brought up to the test temperature with hot air passed through the furnace. After all equipment was up to temperature and air flow adjusted, the dust feeder was started. Next, sample probes were inserted and the test started. Temperatures, flow rate and filter resistance were noted at 10 min. intervals throughout the test period. In most cases, 2 or more upstream dust samples were obtained in succession during the interval required to obtain a significant weight increase on the downstream sample filter (a period of 1 to 2 hr.). Filter efficiency was based on the average dust loading of all upstream samples.

Results

Filter tests were conducted at a number of flow rates, at elevated and room temperature. All test data are summarized in Table II which lists (a) fiber size, (b) filter depth, (c) packing density, (d) filtration velocity, (e) air temperature, (f) dust loading, (g) clean filter resistance and (h) dust removal efficiency pertinent to each test. Dust loadings ranged from approximately 10 to 40 gr./1000 SCF of air. (This is approximately 3½ times the dust loading at 1400°F.)

For ease in appraising the experimental results, data shown in Table II have been rearranged into Table III-VII to clarify the effect of selected variables.

Effect of filter depth on filtering efficiency is shown in Table III for the largest size of fibers at filtration velocities of 400 and 700 fpm. At both flow rates, filtration efficiency increased rapidly when filter depth was increased from 1¼ to 4 in., but successive increments produced less and less improvement. This is probably due to 2 factors: (1) difficulty in achieving good coverage at the edges and in the corners of the filter frame with thin layers of media, (this is especially difficult when working with large diameter fibers because of the relatively few numbers of fibers actually present to fill the voids) and (2) the dust removal by successive increments of filter media is quite different when dealing with an aerosol containing a dust of uniform size and when the aerosol contains a wide diversity of sizes. In the former case, each equal increment of filter material removes the same percentage of dust reaching it. In the latter case, particles possessing sizes for which the filter has high efficiency are generally almost completely removed in the very first sections, whereas particles which are of a size to escape capture in the first layer tend to penetrate all successive layers with little diminution. Therefore, the rate of dust removal tends to decrease rapidly with successive additions to the filter when dealing with polydispersed dusts and deep-bed, high porosity filters. It may be noted from Table III that a 4 in. deep filter composed of the largest diameter fibers gives results very nearly as good as does an 8 in. depth (while a 1½ in. depth of filter is much inferior). Similar patterns of dust removal have been noted elsewhere.[6]

The dust removal efficiency of the 6 in. deep filter at a flow rate of 400 fpm. was greater than that of the 8 in. filter. It was noted that the air flow resistance of the clean, 6 in. filter was also greater. It is believed that these efficiency and resistance measurements indicate that although the 6 in. deep filter contained only ¾ the weight of fibers present in the 8 in. filter, its individual fibers were dispersed more evenly and uniformly and, therefore, the 6 in. deep filter contained a greater number of effective fibers than did the 8 in. deep filter. The experimental filters were all hand packed from large batts of loose, randomly oriented fibers and variations in shot content as well as in the hand packing procedures used by various investigators were noted. For large scale application fibers would be prepared in the form of machine-packed batts of uniform porosity and fiber content and much greater uniformity in performance (from filter to filter) could be anticipated.

Packing density was not one of the filter characteristics specifically studied although small variations were made during the tests to observe the effect on filter stability under high velocity and high heat conditions. In one instance where a comparison may be made (*i.e.*,

TABLE II

Filter Efficiency and Resistance

Fiber Diam. μ	Filter Depth In.	Packing Density[a]	Gas Velocity fpm.	Gas Temp. °F	Filter Resist. in. w. g.	Dust Loading gr./1000 ft.^{3}	Wt. Efficiency %
20	1.5	2	350	1400	0.5	36.5	79
20	1.5	2	700	1400	1.3	22.4	71
						21.4	70
20	4	2	400	1400	0.8	29.4	86
20	4	2	700	1400	1.4	16.3	82
20	4	3	400	70	1.2	9.2	85
20	4	3	400	1400	1.7	20.3	83
						16.0	80
20	6	2	350	1400	1.8	20.8	88
						22.1	89
						27.5	92
20	8	2	400	1400	1.4	35.1	87
20	8	2	700	1400	2.8	14.4	84
8	1.5	3	110	70	0.6	43.2	99
8	1.5	3	350	70	1.7	30.1	99
						26.0	99
8	1.5	3	400	1000	2.2	33.9	94
8	1.5	3	700	1400	—	16.4	84
						22.8	83
4	1	2	110	70	0.5	37.1	99
4	1	2	350	70	2.4	36.8	98
						8.9	98
4	1	2	700	1500	3.7	6.6	85
4	1	2	400	1400	2.3	34.3	91
Composite[b]		3	400	70	5.1	13.1	99+
Composite[b]		3	400	1400	5.3	10.3	99+
						10.8	99+

[a] Shot content: 65% by wt. Packing density of fibers only 35% of values shown.
[b] Composite filter contained 5 in. of 20 μ fibers, 1 in. of 8 μ fibers and ⅜ in. of 4 μ fibers.

TABLE III

Effect of Filter Depth on Filtration Efficiency (20 μ fibers at 1400°F)

Filter depth In.	Efficiency—% by weight 400 fpm.	700 fpm.
1.5	79	71
4	86	82
6	90	—
8	87	84

[6] First, M. W., Moschella, R., Silverman, L. and Berly, E., Ind. Eng. Chem., 43, 1363 (1951).

TABLE IV

Effect of Temperature on Filtration Efficiency

Fiber diameter μ	Filter depth in.	Face velocity fpm.	Efficiency—% by wt. 70°	1400°F
20	4	400	85	82
8	1.5	400	99	94
4	1	400	98	91

4 in. depth of large diam. fibers at 1400°F and 400 fpm. face velocity) the lighter pack showed a somewhat higher efficiency and it is believed (as noted above) that an important factor is the degree of fiber dispersion and uniformity of packing.

Effect of temperature on filtration efficiency is summarized in Table IV. In each instance filtration efficiency decreased with increase in temperature, as predicted by impaction theory. For the examples cited in Table IV penetration increased by factors of 1.2, 6.0, and 4.5 when air temperature was raised from 70° to 1400°F. for the fibers of large, medium, and small diameter, respectively. Although additional observations on aerosols containing particles of uniform size are required to evaluate the exact relationship more adequately, these results appear to be of the right order of magnitude, as predicted by theory.

Effect of filtration velocity is an important factor in dust removal efficiency. When impaction is the primary separation force, theoretical considerations (Equation 2) indicate that efficiency should increase with increases in gas flow rate. Investigations of the dust removal efficiency of 50 to 250 μ diam. glass fibers for spherical and irregular dust particles having a mass median diam. of 2.5 μ (4, p. 80) have confirmed this relationship at flow rates between 100 and 324 fpm. Results obtained in the present study pertaining to the relationship between dust removal efficiency and gas flow rate at 1400°F are shown in Table V. In each case efficiency declined at the higher flow rate, contrary to the prediction of Equation 2 and previous experience with the behavior of glass fibers at room temperature. There is no present evidence to indicate that high temperature per se can be effective in reversing the velocity effect and it was concluded that at the 700 fpm. flow rate separations occurred between the filter frame walls and the filter medium of such magnitude as to more than offset any increase in efficiency that might occur due to an increase in velocity. After prolonged exposure to high temperatures and high air velocities it was frequently noted that the filter pack appeared to be considerably compressed at the center and it is assumed that the contractions at the center caused a corresponding rise at the edges to produce a separation at these points. It is believed that an initial increase in filter efficiency at 700 fpm. caused dust to deposit as a high-resistance layer on the surface of the filter (instead of penetrating throughout the filter depth) and this accelerated the observed results.

Observations of the effect of velocity on efficiency at room temperature were made on the smallest and on the medium diameter fibers, only, at flow rates of 100 and 400 fpm. For these conditions, removal efficiency was very high. No significant differences in filtering efficiency were noted and it may be concluded that at flow rates below 400 fpm. no significant distortion of the filter pack occurred.

Effect of fiber size on dust removal efficiency at high temperatures is shown in Table VII. For low temperature filtration it has been found (4, p. 79) that the finer the fiber the smaller the particle size removed at high efficiency and similar results were obtained at 1400°F.

Resistance of each filter is indicated in Table II. In general, resistance was directly proportional to air flow rate for each of the fibers (both at high and low gas temperature) in accordance with the pressure drop equation for viscous flow. Iberall[8] and others have indicated that viscous flow conditions are present in very porous beds of cylindrical fibers when the Reynolds Number of flow is below approximately 1.2. Calculations covering the full range of these experiments indicated that in all cases viscous flow conditions prevailed. For example, at 70°F it requires 150 fpm. velocity around a 20 μ diam. fiber to give a Reynolds Number of flow of 1, while at 1500°F the required velocity for the same flow number is approximately 1500 fpm.

For the largest diameter fiber (the only one for which data are available) filter resistance was observed to be proportional to filter depth at 400 and 700 fpm. flow rates.

Increases in temperature resulted in increased filter resistance at equal volumetric flow rates but these increases were not in proportion to the rise in the value of the ratio of viscosity to density. As discussed above, resistance increases in conventional units (in. water gage) should be in proportion to the rise in the value of absolute viscosity: a ratio of 2.3 for a temperature rise from 70° to 1400°F. Although increases were noted at 1400°F, in all cases they were less than double the resistance at room temperature and this discrepancy cannot be entirely accounted for by increased separation of fibers from the edges of the metal frame under conditions of high heat.

[8] Iberall, A. S., *J. Res. Nat. Bur. Stds.*, **45**, 398 (1950).

TABLE V

Effect of Filtration Velocity on Efficiency

Fiber diameter μ	Filter depth in.	Efficiency—% by wt. 70° F 110 fpm.	70° F 400 fpm.	1400°F 400 fpm.	1400°F 700 fpm.
4	1	99	98	91	85
8	1.5	99	99	94	84
20	1.5			79	71
20	4			86	82
20	8			87	84

Graded Fiber Filter

Based on the resistance and efficiency characteristics of the 3 individual fiber sizes, a filter was prepared combining all 3 fibers in order to provide high efficiency (small diam. fibers) low air flow resistance (large diam. fibers) and long service life through storage of large amounts of dust without undue resistance rise (large and medium diam. fibers) or dust blow-off (medium and fine fibers). The amount and size of fibers required for this service are dependent in large measure upon the character of the dust to be removed. For the particular aerosol described above the following fiber sizes and amounts were chosen and arranged with the coarsest fibers upstream and the finest downstream.

Fiber Diam. μ	Packing Density (Fibers Only) Lb./Ft.³	Depth of Fibers, In.
20	1.0	5
10	1.1	1
8	1.1	⅛

When tested at 70°F and 1400°F at a gas flow rate of 400 fpm. and dust loading of approximately 10 gr./1000

SCF, dust removal efficiency was, in all cases, greater than 99% by weight and air flow resistance was approximately 5 in. of water gage, (Table II).

The particle size distribution of the dust escaping this graded fiber filter is shown in Fig. 10, Curve C. Based on 99% dust collection efficiency the particle size efficiency of the graded fiber filter is as follows:

Size range μ	Removal efficiency % by weight
0-1	78.4
1-2	95.4
2-5	99.4
>5	99.9

Visual examination of the filter layers at the conclusion of the tests indicated that the mass of dust was concentrated within the 2 coarsest fiber layers and only slight staining of the final fine fiber layer occurred. The fact that the dust was well distributed throughout the fiber mass was favorable for long service life and slow resistance rise with dust deposition. Twenty-five hundred grains of dust/ft.^{3} of filter were deposited in the graded fiber filter during the course of the tests without appreciable resistance increase. Most important, from the standpoint of long service life, the dust did not accumulate at the surface of the filter or at the interfaces between layers of different fiber sizes and form a high resistance dust layer. Although other sizes and proportions of fibers might be required for high efficiency collection without rapid resistance increase with different particle size distributions, information now available serves as a guide to predict the performance of a wide variety of combinations. Typical filter resistance data related to time of exposure and weight of dust deposited are shown in Table VII for each of the uniform fiber sizes and for the composite, graded fiber filter. Although dust exposure was relatively light, the uniform fiber packs and the composite fiber filters indicated adequate dust storage capacity even at high temperatures.

Dust Retention

Viscous liquids are commonly used to coat large diameter fibers (100-250μ) employed for filtering coarse dusts. Their function is to retain captured dust. Viscous coatings are obviously impractical at the temperatures used in this study. Although dust retention is not a problem at room temperature with fibers less than 20 μ in diam., it was necessary to determine whether dust retention properties were altered at high temperatures. This was tested by sampling downstream of a 4 in. deep dust-laden filter after the dust feeder was turned off. Similar tests were made while the filter frame was rapped 40-50 times/min. with a heavy object to simulate an irregular vibration effect. Tests were conducted at 1400°F and air flow rate of 400 fpm. Immediately after the dust supply was stopped, a dust deposit equivalent to approximately 0.5 gr. of dust/1000 ft.^{3} of air was collected on the sample filter, but subsequent samples for periods up to 3 hr. showed complete absence of dust in the air following the filter, even after rapping periods. Whether the dust found downstream of the filter immediately after the dust feeder was turned off represented blow-off from the fibers or was dust which had accumulated on filter frame and duct walls was not definitely determined, but the fact that similar results were obtained with the composite filter, which contained a pad of small diam. fibers on the downstream side to prevent blow-off (as well as to retain small diam. particles) indicates the latter explanation is more probable. In any case, the total weight of dust appearing on the downstream side of the filter after dust feeding had stopped was insignificant in relation to the dust removal performance of the experimental filters.

TABLE VI

Effect of Fiber Size on Filtration Efficiency (1.5 in. filter depth)

Fiber diam. μ	Efficiency—% by wt.			
	70° F		1400°F	
	110 fpm.	400 fpm.	400 fpm.	700 fpm.
4	99	98	91[a]	85[a]
8	99	99	94	84
20	—	—	79	71

[a] 1 in. deep filter.

Fiber strength after prolonged exposure to high temperature and pressure (due to air flow resistance) as well as to vibration and rapid heating and cooling, was determined for each of the 3 fiber sizes. Static tests for pressure resistance were performed by placing a weight of the appropriate size on top of plugs of fibers and putting the assembly in a muffle furnace at 1400°F for a number of hours. Dynamic tests were made by subjecting filters to the effects of hot air flow. In the latter experiments it was possible to add the effects of vibration as well. The results of static and dynamic compression tests were equivalent. Typical results by the static method are shown in Table VIII for pressures equivalent to 4 and 12 in. water gage resistance. In each case there was a small decrease in thickness of the pack under heat and pressure. After the initial compression the filter appeared to be stable, however, and this agreed with observations on test filters subjected to flow rates of 400 fpm. or less. (As noted above, displacement of the filter pack occurred at flow rates of 700 fpm.) It is believed that hand packing results in inhomogeneities which vibration and pressure serve to relieve. Similar results may be obtained by dropping the newly packed filter face-down on a bench a few times to settle the fibers prior to testing. Observations of this nature are also shown in Table VIII.

Filters which were alternately heated and cooled between 1400 and 350°F for 8 hourly intervals showed no detectable changes to fiber or pack other than the initial compression noted above, while static tests indicated excellent fiber stability over a much wider range of temperature extremes.

Chemical resistance of the fiber to the collected dust is an additional consideration. Although this is generally not important at ordinary temperatures, inert substances often become chemically active at high temperature. It was noted at the conclusion of high temperature filtration tests that the *Fiberfrax* had changed color from white to a deep, coppery red. Prolonged treatment of the discolored fibers (15 hr.) with common mineral acids failed to change their appearance and it was concluded that the

TABLE VII

Resistance Rise and Weight of Dust Deposited in Filter

Fiber diam. μ	Filter depth in.	Filtration velocity fpm.	Air Temp. °F	Total dust deposited. gr./ft.^{3} of filter vol.	Filtration period min.	Resistance increase in w. g.
20	6.0	350	1400	820	271	0.05
8	1.5	400	1000	10000	120	0.55
4	1.0	700	1400	4700	115	0.15
Composite	6.1	400	1400	2500	360	0.05

TABLE VIII
Compression Tests on Fiberfrax Fibers
(12 hr. test)

Temp. ° F.	Pressure[a], In. Water Gage	Filter Thickness, In.					
		4-μ Fibers		8-μ Fibers		20-μ Fibers	
		Start	End	Start	End	Start	End
70	0	3	3	3	3	3	3
1400	0	3	3	3	3	3	3
1400	4	1 3/8	1 1/4	1	7/8	1	7/8
1400	12	1	7/8	3/4	5/8	3/4	5/8

[a] Actual stress produced by mechanical compression. Same pressure drop due to air flow probably produces less compression because it is a distributed load. Therefore these results are conservative.

material producing the color was not a film of copper oxide or reduced copper on the surface of the fibers but was, in fact, a change in the character of the surface layer of the *Fiberfrax* itself. X-ray diffraction studies failed to reveal the presence of any crystalline structures in the discolored fibers and it was concluded that the copper oxide was behaving as a flux. Cupric oxide has been used as a flux for ceramics, and similar materials are used as fluxing agents in the regular manufacture of *Fiberfrax*. Even though cupric oxide is only a synthetic test material this point was considered relevant since other materials of this general nature will also act as fluxes. However, a devitrification test of 15 hr. at 1450°F with a used filter did not show embrittlement of the fibers. This indicates that although some combination is taking place between *Fiberfrax* and copper oxide, there is no measurable effect on fiber embrittlement, or strength.

Conclusion

Air cleaning devices in current use capable of receiving hot gases without 1 or more prior cooling stages include: (a) mechanical and inertial separators such as cyclones and settling chambers, (b) wet collectors such as fog- and Venturi scrubbers and (c) electrostatic precipitators. Mechanical and inertial collectors may be constructed from a variety of materials having good heat resistance but are limited in dust removal ability to particles greater than approximately 10 μ in diam. For this reason they are often employed only as precleaners to reduce the bulk dust loading reaching more efficient devices. Wet collectors, also, may be constructed from a number of heat resistant materials and, in addition, have excellent gas cooling capacity because of the evaporation of large volumes of water. Some devices of this kind have high efficiency for particles less than 1 μ in size. In areas where water is scarce or of an unsuitable quality, wet methods may be impossible or uneconomic. Further, permission to dispose of a wet sludge is not easily obtained in many localities. The action of soluble corrosive substances removed from the hot gas stream is often an important consideration in the useful life of wet collectors. Electrostatic precipitators are capable of high efficiency for very fine particles (*i.e.*, less than 0.5μ) at temperatures up to about 600°F. Although gases at temperatures as high at 1100°F have been treated, the electrical resistivity of dry powders and dusts generally becomes excessive at these temperatures. In addition, heat warpage of ionizing and collecting surfaces may occur to disturb the electrical balance of the system.

Industrial bag filters may be used for the cleaning of moderately hot air and gases; the maximum temperature depending on the specific filter medium employed. For example, woven glass-fiber bags have been found suitable for metallurgical gases up to 500°F[1] and Orlon for temperatures up to 275°F[11] but sustained exposure to higher temperatures has been found to greatly shorten bag life. Hotter gases may be precooled prior to reaching the bag house by means of air dilution, heat exchangers, radiant cooling or evaporative cooling. Cooling by dilution with ambient air increases the size and cost of the dust collecting equipment many fold in the case of very hot gases and is clearly uneconomical. Performance of heat exchangers operating on dirty gases has been reported as uneconomical both with regard to initial and maintenance costs[3]. Radiant cooling is effective when the temperature of the gases is very high but for cooling of gases below 1000°F the area of heat exchange surfaces required becomes unduly large.

Evaporative cooling utilizing fine spray nozzles is often employed for this type of service because of the low cost of water and its excellent cooling properties. However, the water required to cool very hot gases materially increases the gas volume to be treated. For example, if 100 ft.^{3} of air were cooled from 2000° to 250°F, air volume would be reduced to 29 ft.^{3} By evaporation, approximately 15 additional ft.^{3} of water vapor would be added in the cooling process and although net volume after cooling would be reduced to less than 1/2, the cooling water vapor would account for about 1/3 of this total. Condensation of corrosive, water-soluble chemicals from warm, moist gases when in contact with the cool outer walls of dust collecting equipment has proved to be an objectionable factor in many installations (even when heat insulation has been provided). Regardless of the method chosen to cool gases prior to treatment in a bag house, elaborate automatic controls are always necessary to prevent overheating or condensation of moisture; either of which leads to destruction of the filter cloth. Therefore, equipment and media suitable for the filtration of hot gases without the necessity for prior cooling have a number of important advantages: (1) reduction in overall size of equipment required, (2) simplicity of temperature and other controls and (3) freedom from condensation and corrosion.

The hot gas filter tests performed in the course of this study for the specific purposes requested by a contractor of the Atomic Energy Commission do not represent a full exploration of all the useful properties or fields of application for filter fibers resistant to high temperatures. However, sufficient information has been presented to indicate that when very high temperature filtration is required the necessary materials are now available. Porous beds of fibers are suitable for low dust loadings but special applications may occur when the cost and labor of frequent filter renewals is offset by the more desirable features inherent in the direct filtration of the hot aerosol. General application to dust loadings in the more usual industrial range (*i.e.*, 0.1 to 10 gr. of dust/ft.^{3}) will probably require a woven fabric possessing sufficient mechanical strength to withstand a large number of filtering and cleaning cycles but novel methods of using heat resistent fibers for the filtration of hot aerosols are under active consideration.

Our test program indicated the manifold difficulties involved in evaluating filter performance at 1400°F. Stainless steel glows at this temperature and simple manipulations such as introducing or removing a sampling probe from the test duct become deliberate and laborious when performed in close proximity to red hot surfaces while wearing heavy, insulated mittens. Multiple sets of sampling and hot test equipment were re-

[1] See footnote 1, page 63.
[11] Pring, R. T., *Heat. & Vent.* (December 1952).
[2] See footnote 2, page 62.
[3] Bainbridge, R., *J. Metals* (Dec. 1952).

quired so that instruments could be set aside for cooling after use without delaying the progress of the experiment. Flow adjustment and meter calibration were complicated by the fact that both temperature and air flow rate affect the response of head-type rate meters. It was discovered that it required 3 to 4 times as long to perform a given test at high temperature as was required at room temperature.

From a relatively few tests on individual fibers it was possible to design a filter to meet specific efficiency, resistance, capacity and life requirements. In addition, these data may be used for the solution of other difficult high temperature dust cleaning problems.

An Apparatus for Quantative Studies of Inhaled Aerosol by Dosimetric Procedure*

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Our knowledge of hazards resulting from exposures to noxious gases and aerosols by inhalation is based on experimental procedures involving characterization of the atmosphere into which animals are placed and observation of toxic effects. Results are in terms of air concentration (mg./m.^{3}, or ppm.), and the physiological or toxicological effect from this concentration. While this method of testing is useful, it has limitations. Variations in respiratory pattern that can affect retention of inhaled vapors or aerosols are not taken into account. In many studies of physiological and pharmacological responses resulting from inhalation of noxious materials it is often necessary to know the exact dose retained in the animal's respiratory system.[1] This information can be obtained by determining the amount of material an animal inhales and exhales. The difference is the retained dose which can be expressed in terms of mg./kg. of body weight. While retention of airborne materials applies to vapor or aerosol, this paper will be limited to a description of instrumentation designed to investigate the toxicity of aerosols. This apparatus, which has undergone various changes and modifications in refinement, can undoubtedly stand further improvement. It was designed primarily for the dog as the experimental animal. The principles, however, are applicable to man. One of the basic considerations in design was the use of equipment for study on unanesthetized, as well as anesthetized animals. Comfort, therefore, became an important factor so several hours' experiments could be conducted. The apparatus consists of a dispersion chamber, a sub-chamber assembly, and the physiological recording instruments.

[1] L. Dautrebande. Physiological and Pharmacological Characteristics of Liquid Aerosols. *Physiol. Rev.*, Vol. 32, No. 2 (1952).

* Presented at the 49th Annual Meeting of the Air Pollution Control Association held in Buffalo, N. Y., May 20-24, 1956.

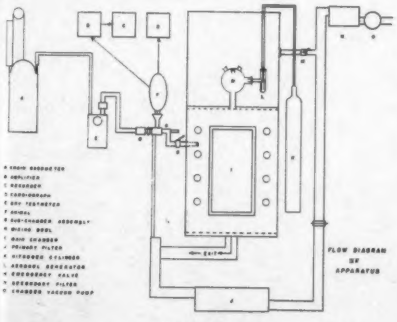

Fig. 1.

Description

(A) Dispersion Chamber

In order to maintain uniformity of concentration and particle size distribution of the aerosol, a dynamic-type chamber of 200 *l.* capacity was used. Fig. 1 shows a schematic flow diagram of the entire apparatus. The aerosol was generated and introduced into the air stream at the mixing bowl. Near the point of entry particulates are mixed with the air stream and enter the chamber via a distribution plate, assuring a uniform concentration in the chamber. The chamber effluent passes through a cross arrangement of perforated tubes through a central exhaust plenum. At the chamber exit the aerosol is removed by means of a standard 600 cfm. particulate and vapor CW collective protector unit. Airflow through this system is controlled by a 1 in. orifice and a 2 in. gate valve situated between the primary and secondary or emergency filter. Filtered air is exhausted outdoors through a stack. A 1½ hp. centrifugal compressor rated at 250 cfm. against a static pressure of 15 in. of water serves as the exhaust pump. A 150 *l.* lucite safety chamber incloses the aerosol generator and the mixing bowl. Additional safety features have been incorporated with this system by having a quick opening valve between the chamber and emergency filter. Release of this valve can flush the system of toxic aerosols at 60 cfm. This device guards against contamination of room air in the event of development of abnormal resistance to airflow through the primary filter.

(B) The Sub-chamber Assembly

The aerosol is pumped from the dispersion chamber through a 1-in. 3-way quick-surging valve into a 1-*l.* inhalation plenum. This valve makes it possible to introduce room air, or aerosol to the inhalation plenum. Airflow in this system is controlled by means of ¼-in. orifice near the primary filter. Thus, chamber and plenum exhausts are parallel. Fig. 2 shows a diagram of the sub-chamber assembly. Directly attached to the plenum is a 2-way demand valve (Fig. 3) which has a dead space of 20 cc. The dog, wearing a special face mask[2] (Fig. 4), inspires from the plenum through one of these valves (M-9-gas mask C-15 outlet valves in phase, inspiration closing the expiration valve). Concentration of the aerosol and particle size distribution are measured from the plenum by means of

[2] J. P. Saunders and R. W. Fogleman. The Respiratory Effects of Inhaled Toxic Vapors in Air I. An Apparatus for Quantitative Measurements of Inspiratory and Expiratory Air. CmlC., *MLRR No.* 179 (1953).

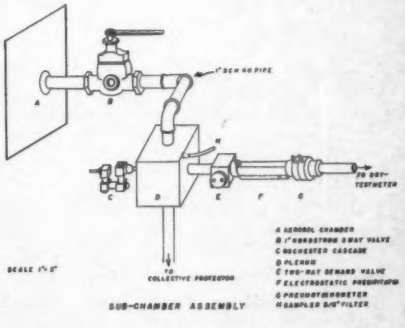

Fig. 2.

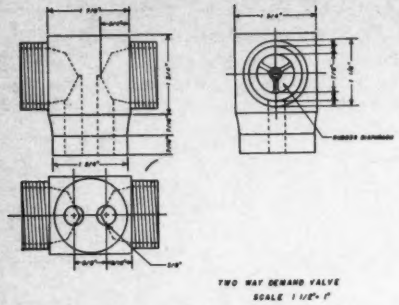

Fig. 3.

a filter paper sampler and a cascade impactor, respectively.

The dog exhales through the second valve through an electrostatic precipitator[3] (Fig. 5) which collects the exhaled aerosol. The exhaled air flows first through a 20-mesh stainless steel screen down-stream to the precipitator and thence to a 1-in. pneumotachometer (Fig. 6) which contains a 400-mesh monel wire screen. The air is then dried through a Drierite bed, fitted on an M-9 gas mask canister which removes the ozone from the effluent stream. Finally, the air passes through a dry test meter and is ultimately collected in a 120-*l.* gasometer.

(C) Physiological Measurements

Any number of recording systems commercially available may be used for the physiological measurements. In this apparatus a 9-channel oscillograph is employed. Strain gages are used as pressure transducers and, when necessary, the circuit from these gages is fed through a carrier amplifier where the input voltage is rectified before passing to the galvanometers in the recorder. Fig. 7 shows records taken from 3 anesthetized beagle dogs showing respiratory effort, tidal volume, blood pressure, and neuromuscular response. The ECG is recorded independently, using a Cardiall.

[3] A. D. Hosey and H. H. Jones. Portable Electrostatic Precipitator Operating from 110 Volts A.C. or 6 Volts D.C., *Arch. of Ind. Hyg., and Occ. Med.*, Vol. 7, No. 1 (January 1953).

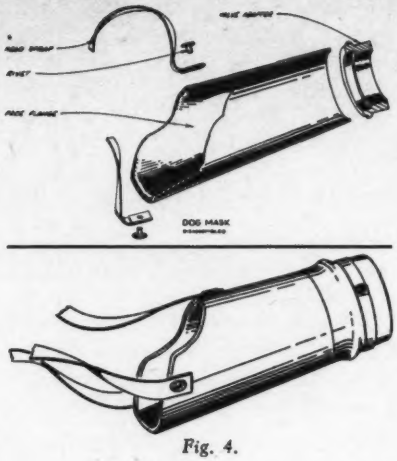

Fig. 4.

Performance

(A) Chamber-Plenum System

Performance characteristics of the chamber-plenum system have been investigated and results are shown in Fig. 8. From this curve one sees that at any given flow rate (200 to 1850 l./min.) 92.5% of the total airflow through the system passes through the chamber proper, the remaining 7.5% passes through the inhalation plenum. These flow rates approximate the ratio of the orifices (1 in. and 1/4 in.) in the system. In accordance with the principles of chamber operations reported by Silver,[5] the plenum system attains equilibrium with the chamber within 2.0 sec. at a maximal flow, and within 18.0 sec. at minimal flow. At the lowest usuable flow rate, therefore, the plenum air is in equilibrium with the chamber within the fourth to the sixth breath of a normally respiring dog. This is based on records such as Fig. 7 and the equilibration time for the plenum shown in Fig. 8. Fig. 9 shows the resistance in mm. of water below atmospheric in the airflow range of

[5] S. D. Silver. Constant Flow Gassing Chambers: Principles Influencing Design and Operation. *J. Of Lab. and Clin. Med.*, Vol. 31, No. 10, October 1946.

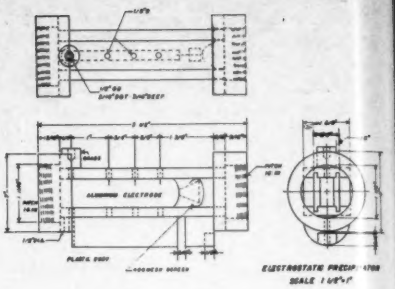

Fig. 5.

the plenum. Much of the work in our laboratories has been done at negative pressures of 6 mm. or less of water (below atmospheric).

Table I shows the results of aerosol distribution studies in the chamber as compared to the plenum at 200 and 600 *l.*/min.

All studies were performed using liquid aerosols of ethylene glycol with a 10% fluorescein tracer. This simplified testing procedures since fluorescein content can be determined by direct reading in a fluorimeter with a known standard.

Data shows that chamber and plenum concentrations were not significantly different at these flow rates. The chamber samples were collected from 10 different positions in the chamber, intermittently, using probe samplers, while the plenum sample was collected continuously. (Some of our recent data shows a trend in plenum concentration slightly less than chamber, at flow rates higher than 600 l/min. The data, however, are too few for a statistical analysis). Fig. 10 shows effects of varying pressure in a pneumatic type generator[6] on concentration in the chamber at a constant airflow of 400 l/min.

(B) The Dosimetric Apparatus

The maximal expiratory resistance in

[6] S. Laskin. Submerged Aerosol Unit. Univ. of Rochester A. E. C. Project. *Quart. Rept. UR38* (1948).

TABLE I

Aerosol Distribution in Chamber vs. Plenum Concentration
10% Fluorescein in Ethylene Glycol

Flow Rate l/min.	No. of Samples	Chamber Concn. mg./m.³	No. of Samples	Plenum Concen. mg./m.³	t.05 df9
200	10	79.2 ± 13.5	1	81.0	no sig. diff.
600	10	26.7 ± 6.6	1	24.0	no sig. diff.

Particle Size MMD = 5.0μ
σg = 2.8

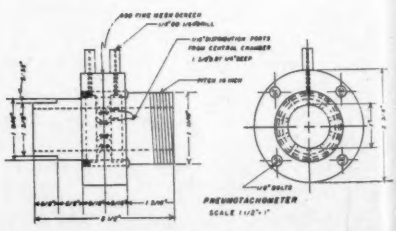

Fig. 6.

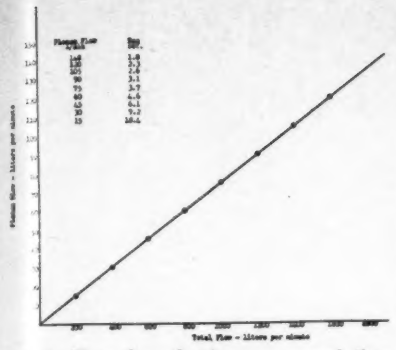

Fig 8. Flow through plenum vs. total flow through system

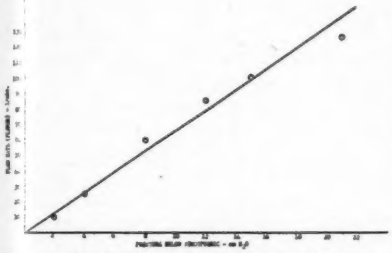

Fig. 9. Inspiratory resistance in plenum at various flow rates

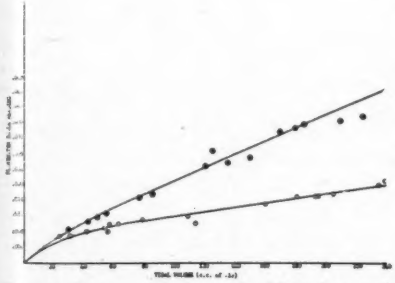

Fig. 10. Pneumatic dispersion of liquid aerosol containing 10% fluorescein in ethylene glycol. Chamber airflow = 400 l./min.

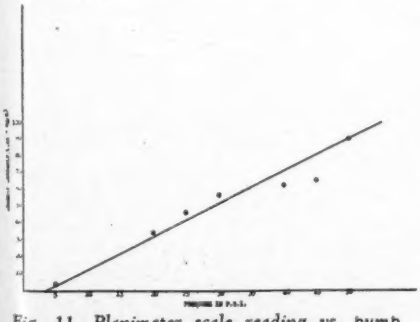

Fig. 11. Planimeter scale reading vs. pump tidal volumes for pneumotach calibration. Attentuation of oscillograph (2)—o. Attentuation of oscillograph (1)—o.

the system is offered by the gas mask canister (1 cm. H_2O at peak flow for dogs at rest). The relationship of pressure resistance developed by the 400 mesh monel wire screen has been worked out by Silverman and Whittenberger.[4] In our system resistance is less than a few mm. of water. Hence a sensitive strain gauge (±0.05 psi.) was used to pick up the pressure drop across the pneumotachometer. Fig. 11 shows the response of the pneumotachometer at comparable pulse airflows at 2 different attentuations on the oscillograph. These calibration curves were obtained using a respiratory pump.

Success of a dosimetric inhalation experiment depends on 2 factors, *i.e.*, the efficiency of the devices used for aerosol collection and leakage in the system. The efficiency of the electrostatic precipitator was tested in 2 ways, by steady state and pulse state sampling. In these tests concentrations of the aerosol were as high as 100-400 mg./m.^{3} and particle size MMD from 1.0 to 8.0 μ. In the steady state, sampling tests at airflows of 30 *l.*/min. (12 KV) the precipitator collected on the average 90% of the aerosol in the gas stream. At lower airflow (3.0 *l.*/min.) efficiency was as high as 99%. In the pulse state sampling tests, efficiency was measured using a respiratory pump. These tests were conducted to simulate dogs breathing at rest using a stroke rate of 20 to 30/min. and a pulse volume of 100-120 cc. The collection efficiency averaged 88% varying from 82 to 95%. In tests designed to determine collection efficiency of a hyperventilating dog (tidal vol.—300 cc. respiration rate 30/min), efficiency dropped as low as 60%. This is one of the critical factors in a dosimetric procedure.

[4] L. Silverman and J. L. Whittenberger. Clinical Pneumotachograph, Methods in Medical Research, Vol. 2, 1950.

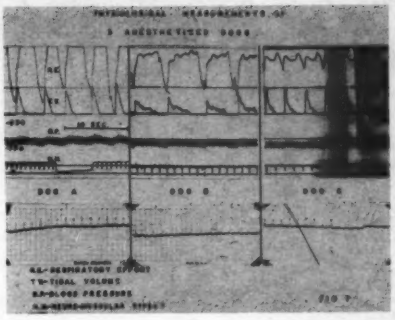

Fig. 7.

To determine leakage in the system 2 dogs were placed on the sub-chamber assembly wearing the special mask. The sole air supply for these animals was from a spirometer (120 *l.* capacity). The animals inspired from this spirometer, through the apparatus, and exhaled into a second spirometer. Table II shows results of this test. Difference of the volumes of the spirometers was taken as a measure of leakage and averaged 2.9%. Whether this is representative of a leak is uncertain. Nevertheless, performance was satisfactory and within limits of experimental error.

Summary

Apparatus for determination of retained dose of aerosols in the respiratory tract of dogs is described. Use of this apparatus provides for the following:

1. Dosimetric studies of inhaled aerosols.

2. Correlation of airborne aerosol (concentration and particle size distribution) with retained dose of inhaled aerosol and toxicological and pharmacological effects (such as respiratory, cardio-vascular, neurological and muscular).

Acknowledgment

The authors are grateful to Mr. Milton Wirth of the Experimental Fabrication Shop, Directorate of Technical Services, CW Laboratories for helpful suggestions in construction of some of the apparatus.

TABLE II

Leakage of Dosimetric Apparatus with Special Mask Using Two Gasometers

Dog	Average RPM	Average Tidal Vol. cc.	Total Inhaled Vol. *L.*	Total Exhaled Vol. *L.*	Loss %	Standard Deviation %
A	9	98	14.19	13.75	3.1	2.2
B	14	80	33.20	32.24	2.7	2.1

The Application of the Concentrometer-Principle To the Determination of Aerosols*

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One of the major problems concerned with the quantitative assay of aerosols is not only to convert all airborne particles at a high sampling rate from the suspended state into a (more or less) 2 dimensional concentrate which renders them accessible to various analytical procedures but to avoid the *denaturation* of the particles by agglomeration, coalescence, and inter-action with each other, *etc.* during the process of forming the concentrate. The method described here represents an effort to avoid, or at least to minimize, these inherent difficulties by permitting for:

(1) a rapid sampling procedure,

(2) the determination of the concentration of particulate matter in aerosols of mutually similar constitution without mass determination,

(3) the microscopic evaluation of the particle-size distribution whereby the danger of *denaturation* of the concentrate is substantially reduced.

The method is based on the principle of the concentrometer [1, 2] by which a graded concentrate of suspended particles is produced by passing the suspension (gaseous or liquid) through a filter material which acts predominantly as a *screen*, i.e. which collects the suspended material quantitatively on its surface. The principle is applicable to the separation from liquids as well as from gases. The following discussion will center on liquids because of certain advantages inherent in the conversion of the aerosol into a liquid suspension prior to its final separation on a surface. The application of the method to the direct filtration of gases will be discussed in a separate paper.

[1] A. Goetz. Theory, Methodology and Application of the Concentrometer, Industr. Assoc. California Inst. of Tech., (Aug. 1955).

[2] A. Goetz. The Concentrometer of Applying Molecular Filter Membranes. J. Am. Water Works Assoc. 45, 933-44 (1953).

* Presented at the 49th Annual Meeting of the Air Pollution Control Association held at Buffalo, N. Y., May 20-24, 1956.

The Working Principle of the Concentrometer

Contrary to the usual practice of filtration whereby each surface element of the filter is passed by an equal fraction V of the total liquid volume, this quantity V varies in the concentrometer along one direction x of the *rectangular* screen element in accordance with a predetermined mathematically-defined function. Hence, if each volume carries the same amount of suspended matter, *i.e.* if the concentration within the liquid is uniform, the concentrate deposited on the screen will represent a *wedge*, the thickness of which varies in a known way in the (x) direction. Equal quantities of fluid of different concentration will thus result in *wedges* of different thicknesses for corresponding positions on the screen.

The working principle of a concentrometer for liquids is represented schematically in Fig. 1, where the liquid (resulting from a sonic impinger) is filled into a container (A), the horizontal cross-section of which is semi-circular. (A) is sealed by a plate (C) and by a resilient thin metal sheet (H). In the center, (C) and (H) have a rectangular aperture.

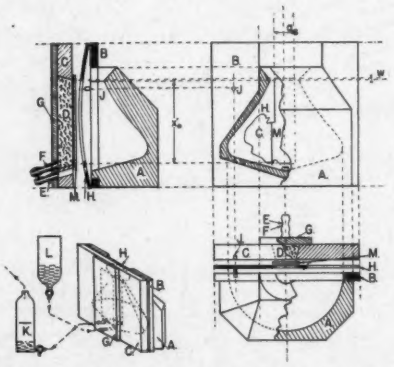

Fig. 1. Design of concentrometer for liquids: (A) cavity block, (B) flange plate of (A), (H) ph.-bronze gasket defining exact screen area, (C) lucite screen holder with insert filled with porous carbon block (D), sealed by plate (G). (J) alignment pins (2). (M) filter membrane. (F) connects with vacuum flask (K). (E) with sample container (L).

In (C) this aperture is filled with a block of uniformly porous carbon (D) which serves as support for the rectangular *screen* (M) which is sandwiched between (D,C) and (H), in a liquid-tight manner, so that (H) masks the exact surface area of (M) exposed to the passage of the liquid in (A).

If by means of a vacuum pump connected with (F) a pressure difference is generated across (M), the liquid in (A) will be forced to flow from (A) through (M-D-F), while simultaneously the liquid level in (A) will be lowered. Consequently the fraction of (M) available to the passage of the liquid will become steadily smaller and the lower parts of (M) will have to filter increasing quantities (V) in order to further lower the level in (A). When the level has reached the lower edge of (M), the fluid remaining in (A) is drained through (E), whereafter (C) is parted from (A) and (M) is removed. (M) carries on its surface the retained concentrate of a density varying along the vertical (x) direction in accordance with a function (V(x)) which is once and for all defined by the geometrical shape of the cavity in (A).

This distribution function (V(x)) does *not* depend on the rate of filtration (*i.e.*, pressure difference or flow-permeability of the screen material), and thus not on a gradual decrease of the flow rate if partial clogging of (M) occurs. The sole requirement is the uniform porosity of (M) at the beginning of the operation. The screen element (M) represents thus a record defining the concentration of the particulate matter previously suspended in the field as it was contained in the defined volume of the cavity in (A). (M) is henceforth referred to as *C-gm.* (concentrogram).

The evaluation of such C-gm. is determined by the following considerations: as the distribution of the flow density through the screen along (x), (V(x)), is always the same, 2 different liquids (of similar nature of suspended matter) with the respective concentrations C_1 and C_2 produce from the same cavity 2 C-gm. represented by:

$C_1(x) = u \cdot V(x)$, $C_2(x) = w \cdot V(x)$, or:

$$\frac{C_1(x)}{C_2(x)} = \frac{u}{w} \qquad (1)$$

While it is physically possible to produce a large (but not infinite) variety of different distributions, $V(x)$, by the suitable design of the cavity shape.

The general mathematical theory of the generation of these distribution functions is somewhat involved and will be published elsewhere. [1]

the selection of a logarithmic (slide rule) function:

$$V(x) = A \cdot e^{kx} \qquad (2)$$

where A, k are fixed instruments constants, is uniquely suitable for this particular purpose.

The principle underlying the evaluation of C-gm. with a logarithmic density distribution is illustrated in Fig. 2 where C_1 and C_2 stem from an unknown, C_S from a known (standard) concentration. It is evident that the shift of C_1 and C_2 against C_S by the distances d_1 and d_2 respectively, juxtaposes surface sections with identical variation of deposit density. This causes between the 2 C-gm. a sharply defined position of equal appearance, demonstrated in Fig. 2 by the precise matching of the lines, for their density (closeness) corresponds to that of the deposit on the C-gm. and varies along x according to eq. (2) and the scale in Fig. 2. In the example the shifts d_1 and d_2 correspond on the scale to:

$$C_1 = 1.67 \cdot C_S,\ C_2 = .67 \cdot C_S, \text{ or}$$
$$C_1 : C_S : C_2 = 1 : 1.5 : 2.5$$

No other but the logarithmic variation of $V(x)$ allows this simple mode of comparison, well-known in photometry. For the deposit density at any distance x_1 along x, eq. (1) results in:

$$\frac{C_1}{C_S} = \frac{e^{k(x_1+d)}}{e^{k} \cdot x_1}, \text{ or}$$

$$\log \frac{C_1}{C_S} = k \cdot d, \text{ or}$$

$$\frac{C_1}{C_S} = \text{antilog}.(k \cdot d) \qquad (3)$$

i.e., the logarithm of the concentration ratio is independent of x_1 and directly proportional to the relative shift d.

The total range, defined by the length of the screen in terms of the corresponding scale, represents in this example the factor of 4, which, for practical purposes, can be arranged to lie between 10^2-10^3, the range depends on the shape and capacity of the cavity A.

The instrument shown in Fig. 1 employs a screen 2 cm. wide and 8 cm. long (.79 in. x 3.16 in.), requires a total volume of 220 ml. and has a range of 200, i.e. the volume which has passed the lowest horizontal strip of 2 mm. (.787 in.) width is 200 times larger than that which has passed the uppermost strip of equal width. It is therefore possible to bridge the entire range of concentrations occuring in aerosol sampling under roughly comparable conditions.

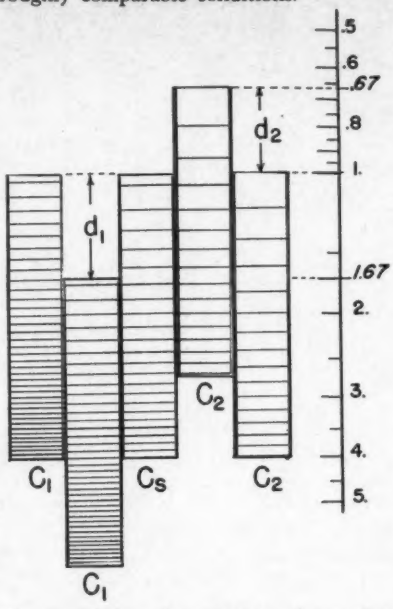

Fig. 2. Scheme of comparison of two (unknown) C-gm. (C_1 C_2) with a standard C-gm. (C_S) of known concentration. The concentrate density which increases downward according to a logarithmic function is represented by the closeness of the horizontal lines. Shifting of (C_1) (left) for the distance (d_1) against (C_S) produces an exact matching of the lines and analogously the shift of (C_2) (right) for (d_2). The distances (d_1, d_2) determined on the instrument scale (right), result in the concentration ratio $C_1 : C_S : C_2 = 1.67 : 1 : 0.67$, the total range in this example being 4.

Properties of Screen Material

In order to realize the method outlined above, the material used for screening must fulfill certain physical and chemical conditions:

(a) the material must have a pore size sufficiently small to retain the smallest particle sizes considered in the assay.

(b) the pore size must be sufficiently uniform, so that the screen, after initial wetting (when the cavity is filled), does not permit anymore the passage of air when the liquid level, while being lowered during the operation, leaves increasing portions of the screen surface to contact with air. This means that the pressure difference required for the filtration may not be larger than the pressure required to overcome the capillary forces due to the surface tension of the liquid within the pores of the filter matrix.

(c) the flow-permeability of the screen must be sufficiently large to effect a complete filtration within a time interval small compared to the settling rate of suspended particles in the fluid.

(d) the screen material must be chemically inert toward the retained particulate matter in order to avoid *denaturation* by reaction between filter substance and deposit.

Filter membranes consisting of stable gels of cellulose esters meet all of these conditions sufficiently well to render the principle of the concentrometer practically applicable.

The development of such membranes goes back to Bechold, Zsigmondy, Elford, Grabar[3] and the work of the Membran-Filter Gesellschaft in Goettingen, Germany. During the last 8 yr. the author and his collaborators continued on this basis and developed membranes which approximate uniformity of pore size and stability in the dry and the wet state for the bacteriological filtration of liquids [4] and of aerosols, the above postulate for a screen-like performance for particle sizes down to 10^{-5}cm. This material is now commercially produced in this country.

The membrane types used for screening impinger fluids permit for a flow velocity of up to 30 cm./min. at 7.4 lb. pressure differential without overcoming the capillary pressure and retaining all particle diam. down to .5 μ quantitatively and a major part (70-90%) down to ca. .3 μ.

These membranes have an additional advantage, as they can be produced with sufficient physical homogeneity to render them almost completely transparent when impregnated with a liquid of nearly equal refractive index ($n = 1.50$) (which must be applied after the complete drying of the membrane, so that the liquid enters the surface opposing the one carrying the retained concentrate). Suitable membrane material can, by careful application of this process, even retain sufficient absence of (internal) light scattering to permit for the application of dark-field condensors of high aperture (cardioid condensor) without serious impairment of image contrast. This ultramicroscopic technique can facilitate the discrimination of particles down to .3-.2 μ. The particular advantage of this procedure consists in a minimal interference with the nature of the individual particle, as is invariably present with the microscopic preparation of the deposit for observation by bright-field illumination because of the requirement of imbedding the object into an immersion fluid to utilize the resolving

[3] P. Grabar. L' Ultrafiltration Fractionnee. (Paris) (1943).
(Excellent reference of earlier work).

[4] A. Goetz. The Application of Molecular Filter Membranes to the Analysis of Aerosols. J. APHA. 43, 150 (1953).
M. W. First and L. Silverman. Air Sampling with Membrane Filters. AMA Archives Ind. Hyg. 7, 1-11 (1953).

power of high aperture objectives. If the particulate matter contains liquid droplets, contact with the embedding fluids can alter their nature substantially by surface interaction or dissolution. This method may prove to be one of the least interfering with the true shape, especially of liquid particles. While the optimal resolving power of the ultramicroscopic technique is, of course, much smaller than that of the electron microscope, its application appears to be definitely superior for liquid particles, since the subjection of the concentrate to shadowing in vacuo, required for an electron-microscopic preparation, leaves serious doubt as to the degree of denaturation inflicted upon the particles.

Conversion of the Aerosol into a Liquid Suspension

One of the major difficulties in the assay of aerosols for particulate matter is the attainment of a sufficiently large sampling rate, because excessive time intervals for the taking of a uniform sample are in general not available. However, in order to obtain a quantity of concentrate sufficient for chemical, *etc.*, analysis a very large air volume has to be sampled, and since the retained matter should be concentrated on as small as possible an area, very high flow densities would be required. A high flow density is in most cases impossible because of the limited flow-permeability of screening materials with sufficient retaining power for the sub-μ sizes occuring predominantly in aerosols. A solution of this problem which (as far as the application to the concentrometer is concerned) has shown definite promise, consists of the quantitative conversion of the aerosol into a liquid suspension prior to the filtration of the resulting liquid concentrate through a suitable membrane. The conversion is effected in as such well-known manner [5,6] by impinging the aerosol onto the surface of a suitable, carefully pre-filtered (membrane) liquid through an orifice under limiting flow conditions, *i.e.*, where the pressure differential exceeds ca. 8 lbs., so that the flow approaches sonic velocity. The author has found it useful to employ slit-like, rather than capillary, orifices for this purpose, apparently because the liquid surface area impinged upon by the aerosol is much larger than for orifices of circular cross-section, and thereby the probability of interception of very small particles by the liquid. Fig. 3 shows

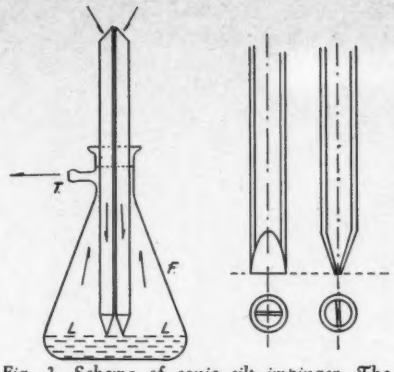

Fig. 3. Scheme of sonic silt impinger. The filter flask (F 2l. capacity) carries the impinger fluid (250-300 ml.) up to the level (L). One or several parallel tubes are placed in the neck of (F) each one ending at the lower end in a narrow slit (.006/8 in. wide, .25/.5 in. long;) The vaccum is connected at tube (T) of the flask, the pump capacity must be sufficient for causing a pressure differential better than 7.5 lb. (½ at.) across the slits.

schematically the assembly of such an impinger unit.

In this manner it is quite possible to attain sampling rates exceeding .1 cbm./min. (3.5 ft.³/min.) in a vol. of impinger fluid of approximately 300 ml. If this quantity of impinger fluid is filtered in the usual manner through a horizontal filter membrane (ca. 10 cm.²), a concentration rate of 10^1/cm.²/min. results which is about 3 times higher than the rate of the customary high volume sampler, and about twice as large as that of direct air filtration through a membrane, applying the same pressure differential. Actually, the concentration rate in a sonic liquid impinger can be increased substantially above this value for the same liquid volume by the parallel operation of a larger number of limiting slit-orifices. The upper limit is the volume capacity of the pumping system applied.

Another advantage of this intermediary conversion consists of the possibility of using different impinger fluids, *e.g.*, water, for the elimination of the water-soluble components or hydrocarbon fluids for the elimination of particles soluble therein, while preserving those of water-soluble nature.

The choice of impinger fluids is limited to those which are neutral toward the substance of the membrane, *i.e.*, celluloseesters, such as water and saturated hydrocarbons, but exclude alcohols, esters, ketones and the like.

Care has to be taken to avoid reactions among the particles and with the fluid. Agglomeration can be avoided by the addition of protective colloids (traces of gelatine, *etc.*), certain reactions can be inhibited by the addition of buffering salts, *etc.*

Evaluation of C-Grams

Determination of the Concentration

The concentration C in *μrg./cbm.* of the aerosol and *c in μgr./ml.* of the impinger fluid is related to the volume of air, V_a: *in cbm.*, and of the fluid in the impinger, V_i: *in ml.*, which latter must be measured after the termination of the impinging period (to exclude losses by evaporation) by:

$$C = c \cdot \frac{V_i}{V_a} = c \cdot \frac{V_i}{t \cdot F} = c \cdot \alpha \qquad \left(\alpha = \frac{V_i}{V_a}\right) \qquad (4)$$

where *t in min.* is the duration of the flow, and *F in cbm./min.* the flow rate, if V_a is determined by a flow meter.

The above relationship permits for approximately similar aerosols the determination of C in the following simple manner: A standard C-gm. is produced from a representative aerosol (of unknown concentration C_s on a tared membrane strip, and the weight increase M is determined by a sufficiently sensitive balance which results in:

$$\frac{M}{V_c} = c_s = \frac{C_s}{\alpha_s} \qquad (5)$$

where V_c is the fixed volume of the impinger cavity ($V_c = 220$ *ml.*).
Hence c_s is the coefficient characterizing the standard. Any C-gm. from another filtration yields, by comparison with this standard, a scale reading, which represents the shift *d* as a proportionality factor (δ = antilog ($k \cdot d$) (see eq. 3). Hence:

$$C = \delta\,(C_s \cdot \alpha) \qquad (6)$$

For example: a C-gm. was obtained by passing 5 cbm. of air through 240 ml. of fluid *(i.e., $\alpha = 48$)* which resulted by comparison with the standard in a shift corresponding to a value on the instrument scale: $\delta = .28$. This standard had a deposit weight of M = 8.2 mgr. (C_s = 37.3 ugr./ml.), hence:
$C = .28 \times 48 \times 37.3 = 500$ *μgr./cbm.*

In this manner it is possible to obtain without resorting to weighing but once, a quite accurate and rapid determination of numerous circular aerosol samples as well as permanent records of the concentration.

The appearance and evaluation of C-gm. is illustrated in Fig. 4 which represents a photograph of 7 C-gm. resulting from soot-containing fluids (as most suitable for photographic reproduction), mounted with the instrument scale (right) with shifts, representing the degree of dilution to which the fluid was subjected. The dilution factor was in all cases 2, except for one, in which it was 4. It is obvious that the juxtaposed sec-

[5] L. Greenberg and G. W. Smith. U. S. Bur. of Mines Rept. 2392 (1922), 3360 (1937).
[6] A. Goetz. Basic Problems in the Detection of Microbiological Air Pollution. *Am. Ind. Hyg. Assoc. Quart.* **16**, 113-19 (1955).

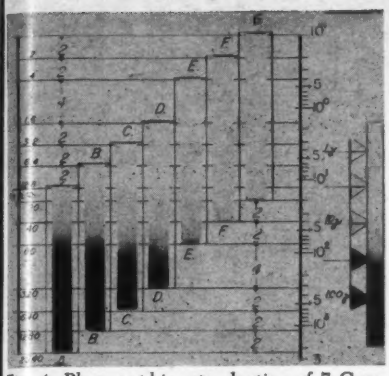

Fig. 4. Photographic reproduction of 7 C-gm. resulting from soot-containing impinger fluid in dilution steps of 2^n (for $0 \leq n \leq 8$ without 2^7), resulting from the concentrometer in Fig. 1. The C-gm. are mounted with relative shift of their reference marks (top) as derived from the scale for the applied concentration ratio. The criterion for the correct realization of a log.-distribution is the equality of gray-shade of their overlapping sections—which appears well met. The C-gm. for the mass-density relation (μ-scale is mounted on the right.)

tions of the C-gm. show the same degree of blackening, corresponding to the mutual shift.

The surface concentration of the retained matter on the C-gm. itself is determined as follows: if c, the concentration of an agent in the impinger fluid is known, *i.e.*, by M eqs. (4, 5), the value of V indicated by the instrument scale for any section of the C-gm. is proportional to the deposit density in this section by the following relationship: If, *e. g.*, ($c = 1.5$ mgr./ml.), the section of the C-gm. at ($V = 7$ ml.) on the scale carries a deposit density of:

$$D = \frac{V.c}{a} = \frac{7 \times 1.5}{.4} = 26.2 \text{ mgr./cm.}^2 \quad (7)$$

where a is the screen area to which the scale refers (in the instrument described $a = .4$ cm.^{2}).

This property of a C-gm. can be used for establishing a correlation of *e.g.*, the color, light-scattering, *etc.*, properties of the concentrate with its density by applying photometric methods for the C-gm. after it has been rendered transparent (see Sec. 3).

Determination of Particle Size and Number

As already stated, the deposit density on the screen can influence the interpretation of the particle size distribution due to reactions within the concentrate, such as coalescence and agglomeration, and lead thus to a misjudgment of the average particle size and number. The graded density of the concentrate on the C-gm. can overcome this difficulty because by a microscopic survey (light or dark field) along the x-direction it is almost always possible to find a region of deposit density where the number of particles appearing in the field area are sufficient in number for statistical confidence, but not dense enough for having been subject to coalescence. This can easily be verified by comparing an area of low, with one of higher density, as a decreasing frequency of small particles in the ranges of higher density is invariably a sign of coalescence.

The determination of the particle number/unit vol. on a C-gm. is based on the following procedure: in accordance with the condition of minimal occurence of coagulated particles a section of the C-gm. is selected for microscopic evaluation. This section is correlated with the V-value corresponding on the instrument scale, *i.e.*, the flow density to which this part of the membrane has been subjected during filtration; then the micro-optical field area a_1, determined with an object micrometer or calibrated reticule, defines the fraction $\frac{a_1}{a}$ (eq. 7) of the volume V which comprises the area for the microscopic count. Hence, the number $\mathcal{N}$ of particles/cbm. of aerosol results from the number n counted in a_1 as:

$$\mathcal{N} = \frac{n}{V} \cdot \frac{V_1}{V_a} \cdot \frac{a}{a_1} = \frac{n}{v} \cdot \gamma \quad (8)$$

where $\left(\gamma = \frac{a}{a_1}\right)$ is a constant for a standardized micro-optical procedure.

The following numerical example may serve as an illustration: an average of ($n = 65$) of particles were counted in a number of squares, each with an area ($a_1 = 2.5 \times 10^{-5}$ cm.^{2}., hence $\gamma = 1.6 \times 10^4$) of the reticule, on a C-gm. section corresponding to ($V = .5$ ml.) which latter resulted from ($V_a = .25$ cbm.) of an aerosol passing through ($V_1 = 250$ ml.) impinger fluid (hence $\alpha = 10^3$), in accordance with eq. (8):

$$\mathcal{N} = \frac{65}{.5} \times 1.6 \times 10^4 \times 10^3 = 2.1 \times 10^9/\text{cbm.} \quad (8a)$$

If C, the mass concentration of the aerosol, has been previously determined on the C-gm. (s.a.), and the average specific gravity g of the suspended matter is approximately known, an estimate of the average particle (as spheres) diam. d can be obtained by the following argument: the total volume of all particles being:

$$U = \frac{C}{g} \times 10^{-9} = \frac{\pi}{6} \cdot \mathcal{N} \cdot \overline{d^3}; \text{ or } \overline{d^3} = \frac{1.9 \cdot C}{\mathcal{N} \cdot g} \times 10^{-9} = \frac{1.9 \times 10^{-9}}{\gamma} \cdot \frac{C \cdot V}{n \cdot g \cdot \alpha} \quad (9)$$

This results in the above numerical example, assuming ($g = 1.2$ and $C = 260$ mgr./cbm.):

$$\overline{d^3} = \frac{1.9 \times 260 \times 10^{-9}}{2.1 \times 10^9 \times 1.2} = 1.96 \times 10^{-18}; \text{ or } \overline{d} = 5.8 \times 10^{-6} \sim .6\ \mu \quad (9a)$$

This average value of d is valid only under the assumption that all particles are of the same size, the result may thus be extremely misleading. One can arrive at an improved estimate by counting in addition to n also n_1, the class of largest particles (*e.g.*, $> 1\ \mu$), and estimate for this class an average diam. d_1. The average diam. d_2 of all other particles results then from eq. (8, 9) as:

$$\overline{d^3}_2 = \frac{n \cdot \overline{d^3} - n_1 \overline{d^3}_1}{n - n_1} \quad (10)$$

If in the above numerical example only 2% ($n_1 = 02$ n) of all particles were found to be of an average diam. of ($d_1 = 2\ \mu$), the average diam. is reduced to almost half of that resulting in eq. (9a), *viz*:

$$\overline{d^3}_2 = 1.96 \times 10^{-18} - 2 \times 10^{-2} \cdot 8 \times 10^{-18} = 36 \times 10^{-18}$$

or:

$$\overline{d}_2 = 3.3 \cdot 10^{-6} \text{ cm.} \sim .33\ \mu \quad (10a)$$

By such successive estimates it is possible to obtain information about averages of particle sizes not any more clearly determinable in the ultra-microscope.

The comparison of C-gm. obtained from the same aerosol but by different impinger fluids (oil and water) yields information about the degree to which the impinger fluids may have affected the particle. Compared with these may be an analogous concentrate on a membrane exposed (by the customary procedure) to direct filtration of a vol. V of air so that the resulting density on it corresponds to that of the region V on the C-gm. selected for the evaluation of the particle-sizes and numbers. An analogous concentrate density is obtained for the membrane area $\mathcal{N}$ used, if:

$$\overline{V} = \frac{V \cdot \mathcal{N}}{a \cdot \alpha} \quad (11)$$

where a characterizes the C-gm. to be compared (eq. 4).

However, the *dry* filtrate represents not necessarily a more accurate picture of the constitution of the airborne matter than the analogous concentrates from impinger fluids, because the particle-size distribution can be (especially for liquid droplets) significantly altered by the retention on, or wetting with the dry membrane surface.

The effect of the concentrate density on the microscopic particle pattern is

illustrated in Fig. 5: two C-gm. of different fluids are mounted together with the V-scale of the instrument used. Of each 3 narrow strips were excised, corresponding to (V = .2,. 2,. 20 ml.), and mounted as transparent specimens on microslides. The resulting microphotographs have the same magnification for the same C-gm. The left C-gm. (A) was photographed with bright-field, the right C-gm. (B) with dark-field (cardioid) illumination. The scales indicate the magnification used.

Application to Molecular Components.

Aside from the determination of particulate matter, the concentrometer permits, in principle, also the measurement of specific molecular components which occur in minute concentrations in the air. To attain this the membrane has to contain within itself a uniformly distributed reagent which reacts specifically with a particular component. Fortunately the peculiar internal structure of the membrane matrix provides for sufficient space among the pores wherein suitable reagents can be imbedded (prior to filtration) in sufficient quantity without impairing the flow-permeability seriously. In order to remain within the matrix during the filtration, the reagent must be virtually insoluble: in order to be still capable of reacting with the molecular component of the fluid during its passage through the matrix, the solubility product of the reagent must be larger than that of the subsequent reaction product. These conditions are most unfavorable in view of the high reaction rates required because of the short time (ca. 3 x 10^{-3} sec.) of contact available to the passing fluid. However, this is compensated by the large number of pore-openings in the surface of the membrane (ca.10^8/cm.^{2}) which *subdivide* the flow to the degree required for a high reaction efficiency. Preferably a reagent is applied which produces a distinct color change by the reaction with the trace component in the fluid. As the intensity of this color signal depends, at least in a narrow range, on the number of trace molecules passing through the membrane, *i.e.*, on the flow-density, a C-gm. with a uniform distribution of the reagent will show after filtration a characteristic color variation, the location of which indicates the concentration of the specific component in the fluid.

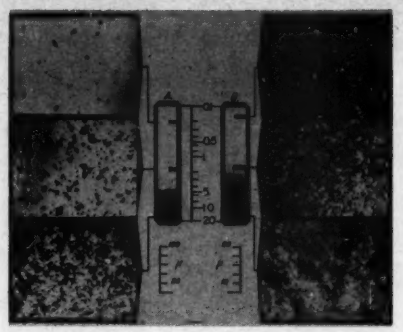

Fig. 5. Photographic reproduction of 2 C-gm. from different impinger fluids with corresponding scale. Three narrow horizontal strips were excised out of (A) and (B), corresponding to the V-values: .2, 2.0, and 20 ml., representing thus an increase in concentrate density of 1 : 10 : 100. The microphotographs obtained from these sections are in bright-field illumination for (A) and dark illumination for (B) and are reproduced with the corresponding size scales. The effect of the concentrate density on the particle size distribution, due to coalescense, is evident.

While this type of application of the concentrometer principle is still under preliminary study it has proven very sensitive and applicable for a few particular cases, *e.g.*, metal ions. For aerosol components which are not soluble in the impinger fluid the procedure is not applicable.

Acknowledgment

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Methods of Generating Solid Aerosols*†

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Production of representative concentrations of solid, liquid and gaseous contaminants in air is a fundamental requirement of research in aerosol behavior and methods of air pollution control. The following discussion deals with generation of solid aerosols. Liquid aerosols may also be generated by some of the methods to be described; ordinary gas mixtures are usually not difficult to establish. Concentrations of solid particles in air are used in laboratory investigations of sampling methods and analytical procedures, or for calibration of sampling equipment and determination of its collection efficiency. Testing of air pollution control equipment (filters, washers, electrostatic precipitators, *etc.)* is undertaken by most manufacturers or by independent laboratories with aerosols made from materials which duplicate the expected service, for design and development information and for final acceptability. Many phases of air pollution research require the simulating of an atmosphere or environment in the laboratory to determine the properties and interactions of the various contaminating components.

Among the allied fields of research which also use aerosols for similar purposes are: chemical engineering (various unit operations involving aerosols); industrial hygiene (industrial air analysis); meteorology (cloud physics); respiratory physiology (lung retention and fate of respired particulates); and military science and engineering (screening smokes and propagation of chemical and bacteriological weapons). Important contributions on the behavior of aerosols and operation of aerosol collection mechanisms continue to be made by investigators in all of the above fields.

Selection of a method of generation will be determined to a large extent by the subsequent use of the aerosol. For example, when low dust concentrations are desired (as might be found in ambient atmosphere) the particular methods chosen will be different from those used for high concentrations (as might be found in industrial dust collecting systems). Methods for production of homogeneous aerosols (for fundamental research) have definite limits as opposed to those producing heterogeneous systems (as usually occur naturally).

Static dust concentrations are produced in containers (as small as a 20 l. carboy) or cabinets (as large as 30 or more m.^{3}) of known (constant) volume by dispersing into them a certain amount of solid material for a short time, and then withdrawing a sample of the aerosol from the static chamber. These are usually used for sampling instrument and procedure calibrations and for

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TABLE I

Materials Used For Production of Solid Aerosols (Condensed, See footnote 49, page 78)

Material	Size Range μ	Approx. Sp. G.	Shape	Remarks
A. Standard Test Dusts				
Air Filter Institute	1 - 80	2.0	Irregular	Ariz. Road Dust Plus Carbon Black and Cotton Linters
Army	1 - 80	2.5	Irregular	Ariz. Road Dust
ASHAE	1 - 80	—	Irregular	Fly Ash Plus Carbon Black
Atmospheric	0.5	1.5	Irregular	Silicates, Carbon Organics, etc.
British	1 - 100	2.5	Irregular	Mineral Dust (Under Revision)
National Bureau of Standards	0.5	2.2	Spher.-Irr.	Fly Ash
Bureau of Mines Respirator			Irregular	
Silica	0.6	2.6		Free Silica
Lead Fume	0.1	8.0	Irregular	Combustion of Lead Tetraethyl
Magnesium Oxide	0.6	3.6	Irregular	Combustion of Magnesium Ribbon
Chromic Acid Mist	1 - 5	2.7	Spherical	Electrol. Acid
Lead Paint Mist	1 - 10	6.6	Irregular	Spray Paint
B. Resuspended Pulverized Materials				
Aluminum Oxide	5 - 40	4.0	Irregular	Abrasive Grits
Asbestos	1 - 5	3.1	Needl-Irr.	Floats (Filter Aid)
Bismuth Subcarbonate	4.0	6.9	Irregular	—
Calcium Carbonate	3.0	2.7	Rhom-Crystal	Ground or Precipitated
Calcium Phosphate	0.7 - 4.3	2.3	Irregular	—
Carbon Black	0.09	1.77	Spher.-Chains	Lamp Black
Carbon Dust	1 - 5	2.0	Irregular	Attr. of Charcoal
Copper Sulfate	1 - 5	3.6	Crystal-Irr.	—
Copper Tartrate	1 - 5	—	Irregular	—
Copper Oxide	4.3	6.4	Irregular	—
Dowex Resin	0.05 - 0.1	—	Spherical	High Static Charge
Fly Ash	16	—	Spher.-Irr.	From Cottrell Precip. of Pulv. Fuel
Glass	1 - 5	2.6	Irregular	Ground Cullet
Glass	7.3	2.6	Spherical	Pavement Marking Beads
Iron	3 - 8	7.8	Spherical	Carbonyl
Iron	0.5 - 6.6	7.9	Spher.-Irr.	Powder
Ilmenite	4.3	4.7	Irregular	Ground Ore
Lindox	0.1 - 0.3	—	Irregular	Zinc Oxide Pigment
Lead Dust	2.2	11.3	Irregular	—
Lithopone	1 - 5	4.3	Irregular	Zinc Sulfide and Barium Sulfate
Magnesium Silicate	2.8	3.3	Irregular	—
Metals	—	1.5 - 10	Spher.-Irr.	Disintegrated Metal Powders
Minerals	—	1.5 - 10	Crystal-Irr.	Ground
Methylene Blue	4.9 - 11	1.26	Irregular	—
Pigments	—	—	Irregular	Metals and Oxides for Paints
Plastics	1 - 5	—	Spher.-Irr.	Molding Powders-High Static Charge
Talc	2.5	3.3	Needl-Crystal	Micronized
Titanium Dioxide	1 - 5	3.9	Spher.-Crystal	Anatase
Silica	1 - 3	2.6	Crystalline	Ground
Silica	0.5	2.2	Spherical	Vaporized-Amorpheus
Sodium Bicarbonate	0.5	2.2	Irregular	—
Sodium Sulfate	4.8	2.7	Irregular	—
Starch	1 - 5	1.5	Irregular	—
Uranium Oxide	1 - 5	—	—	—
Zinc Cadmium Sulfate	2.0	4.0	Irregular	Fluorescent Pigment
Zinc Silicate	1.5	3.7	Irregular	Fluorescent Pigment
C. Solids From Sublimation, Vaporization and Condensation				
Ammonium Bifluoride	0.5	1.3	Spherical	Sublimed from Hot Plate
Ammonium Chloride	0.2 - 10	—	Spher.-Irr.	Controlled Vapor and Condensation
Aluminum Chloride	0.6	2.1	Spherical	Sublimed
DM & DA Smokes	0.2	—	Crystalline	Vaporized
Metals	—	—	Spherical	Aluminum, Brass, Copper, Lead, Sprayed From Metallizing Unit.
Microcrystalline Wax	1.0	—	Spher.-Irr.	Controlled Vapor and Condensation
Rosin	0.2 - 10	—	Spherical	Controlled Vapor and Condensation
Stearic Acid	0.1 - 1.0	—	Spherical	Super-cooled Liquid
Triphenyl Phosphate	0.1 - 1.0	—	Spherical	Super-cooled Liquid
D. Solids From Solutions or Suspensions				
Copper Sulfate	0.3 - 2.7	3.6	Spherical	Spray Solution and Fuse
Copper Tartrate	0.5 - 1.0	—	Irregular	Spray Suspension
Methylene Blue	0.2 - 2.0	1.26	Spher.-Irr.	Atomized Solution
Phenol Red	1.6 - 3.8	—	Irregular	Nebulized Solution
Silver Iodide	0.3 - 0.8	5.7	Irregular	Atomized Solution
Sodium Bicarbonate	0.2 - 0.4	2.2	Irregular	Atomized Solution
Sodium Chloride	0.4 - 1.2	2.2	Cubic-Spher.	Atomized and Fuse
Sucrose	0.03	1.6	Iregular	Atomized Solution
Uranine	1.3	—	—	Atomized Solution
Uranium Oxide	0.5 - 1.7	10.9	Irregular	Spray Suspension
Uranium Trioxide	0.8	7.3	Spherical	Atomize Nitrate Solution and Fuse
E. Solids From Flame Passage, Combustion, or Chemical Reaction				
Ammonium Chloride	0.1	—	Irregular	React Amonia and Hydrogen Chloride
Carbon Black	—	—	Spher. Chains	Comb. with Insuf. Oxygen; Acetylen, Benzene, Butane, Gas
Minerals	—	—	Spherical	Flame Passage of Ground Minerals
Metals	0.001 - 0.1	—	Cub-Spher.-Irr	Direct Comb. or Flame Passage of Powders or Thin Ribbons.
Metals	0.001 - 0.1	—	Spherical	Comb. of Liquid Forms; Lead Tetraethyl, Iron Carbonyl, etc.
Phosphorous	0.1	—	Spher.-Irr.	Screening Smoke From Comb. of White Phos.
Tobacco	0.5 - 1.0	—	Spherical	Comb., Primarily Liquid Droplets with Tars.
F. Biological Materials				
Albumin, Egg	0.5 - 10	—	—	—
Bacteria:				Nebulize
E. Coli	1 x 0.7	—	Cylindrical	—
S. Ind.	0.5 x 1.0	—	Cylindrical	—
S. Marc.	0.6 x 1.0	—	Cylindrical	—
B. Glob.	0.8 x 1.5	—	Cylindrical	—
Bacteriophage	0.02 - 0.05	—	Spherical	E. Coli-T-3
Blood Cells	7.85	—	Disc	Very Uniform Size
Lycopodium	25 - 40	—	Pyramid	—
Pollen:				
Ragweed	19.6	—	Spherical	Ambr. Trifida
Orchard Grass	31.0	—	Elliptical	Dactyl. Glom.
Corn	100.0	—	—	M. Zea.
Rye	55.0	—	—	Secal. Cer.
Spores:				
Pill. Caries	17.0	—	—	—
Bov. Plumbea	5.6	—	—	—

TABLE II

Characteristics of Units for Generation of Solid Aerosols (Condensed)[49]

Name	Feeder Principal	Aeration Method	Classification Method	Feed Rate/Min.	Remarks
Air Filter Institute	Moving Horizontal Tray With Levelling Gear	Venturi Throat	None	1 gram	Fly Ash
ANP	Tube Elevated by Hydraulic Piston into 90 psi Air Line	Sonic Jet	Settling Chamber	1 - 75 grams	Copper Oxide
Atkins	Screw Feeder to Recircl. Unit	Ejector	Recircl. by Fan	8 grams	Pulv. Coal
Bu. Mines	Fluidized Bed from Solids Feeder	Fan	Cyclone	26 pounds/ft.^{3}	Pulv. Coal
Bu. Mines	Tube Elevated by Winch Into Spiralling Air Stream	Ejector	Cyclone or Settling Chamber	Milligrams	Respirator Test Dusts
Bu. Standards	Rotating Gear with Tooth Reduced to Pitch Circle. Grooves Fed by Gravity from Hopper	Ejector	Baffle	1 gram	Fly Ash
Colorado	Tube Elevated by Screw, Dust Scraped Into Spiralling Air Stream	Ejector	Plenum and Filter	Milligrams	Aluminum Hydroxide Therapy
Dust Shaker	Oscillating Vibratory Screen, 40 to 60 Mesh	Air Stream	None	Low	Toxicological Studies
Farr	Four Grooved Plate Moves Horizontally with Levelling gear.	Four Ejectors	None	5 grams	Fly Ash
Geyser	Powder in Small Chamber, Dispersed by Air Jets	Aspirator	None	4 grams	Lithopone Egg Albumin, Kados
Harvard No. 1	Air Dispersed From Vibrating Tube	Air Stream	Settling Chamber	Milligram	Silica
Harvard No. 2	Revolving Disc Screen From Vibration Feeder	Air Jet Under Disc Blower Inlet Above	Elut. Column	grams	Talc Glass Sph.
Harvard No. 3	Rotating 300 Mesh Screen Cylinder, Vibrated, Inside Lucite Container	Aspirator From Bottom Tube	None	1000 particles	Pollen
Harvard No. 4	Rotating Turntable With Scraper, Fed From Vibr. Feeder	Ejector	None	pound	Most dry dusts
Harvard No. 5	Small Screw Feeder, Variable Discharge Openings. To Moving Belt With Rotating Scraper to Adjust Height, Width Fixed by Belt or Notches In Scraper	Ejector	None	300 milligrams	Flake iron powder
Haskell Laboratory	Pulsating Air Flow Through Material in Tube	Air Stream	Three Elut. Columns	—	—
KAPL	Rotating Steel Wire Brush Held Against Aluminum Metal	Air Stream	Impingement Plate	Micrograms	Aluminum or other solids
Minnesota No. 1	Rotating Disc With Dust Ribbon	Venturi Throat	None	0.1 - 2 grams	Fly Ash
Minnesota No.2	Moving Horizontal Trough With Level Dust Layer	Venturi Throat	None	Same	Fly Ash
New York State No. 1	Hopper Feed to Moving Belt, Dust Discharged From End into Hopper With Air Flow Through	Air Stream	Two Imping. nozzles, plus four Elut. Columns	0.1 - 2 grams	Mineral and org test dusts
New York State No. 2	Hopper Feed to Vibrating Chute	Ejector	Impingement Plate	3 pounds	Most dry dusts
Research Cottrell	Funnel Feed To Plate In Horiz. Fan Impeller	Air Stream and Fan Blades	None	—	Fly Ash
Research Products	Compr. Air Jet in Flask With Dust on Bottom	Air Stream	None	0. 1- 2 grams	Fly Ash
Rotating Drum	Rotating Drum With Friable Material. Air Stream Passes Through	Air Stream	None	Milligrams	Charcoal
SRI	Moving Horizontal Vee Trough	Glass Aspirator	Settling Chamber	Milligrams	Carbon black; Powders
Screw	Variable Speed Stoker Screw, Or Wood Auger ½" to 2"	Ejector	—	—	Most dry Powders
Sonic	High Freq. Vibration From Speaker Cone Directed At Dust	Air Stream	—	Low	Many dry Powders
Texas	Modified Bu. Mines Resp. Test Dust Feeder	Ejector	Cyclone	—	Antimony; Sulfur
Univ. Chicago No. 1	Vibrated Sintered 40 Mesh Glass Filter, Air From Top to Bottom	Ejector	None	Milligrams	Similar to Dust Shaker
Univ. Chicago No. 2	Multiple Impinging Nozzles Fed From Dust in Container	Air Stream	Series Impinging Nozzles	Micrograms	
Vibratory	Trough or Tray Pulsed By Electromagnetic Vibrator	Ejector	—	Grams-pounds	Many dry Powders
Wright	Rotating Packed Drum Moved Against Scraper, Dust Entrained in Low Pressure Air Stream	Ejector	None	Micro-Milligrams	Pulv. Coal; Silica & Lead Dusts

physiological studies. Dynamic concentrations are established in moving air streams by continuously dispersing constant amounts of material over long time periods. (A chamber may also be used for mixing or elutriation of dynamic concentrations to produce more uniform aerosols.) Continuous generation of aerosols is used for testing of most dust collection devices, for fundamental studies of aerosol behavior under the action of collecting forces, for research with larger particles which rapidly settle out of static concentrations, *etc.* It is apparent that generation methods for static and dynamic systems may be quite different.

Redispersion Methods for Generation of Solid Aerosols

Redispersion of Dry Powders

Redispersion of dry powders is probably the most widely-used of all methods for producing solid aerosols. Finely-divided materials with a wide range of particle sizes, shapes, specific gravities, and surface characteristics are readily obtainable. A list of powdered materials which have been used as aerosols is given in Table I, A and B. (Tables I and II are condensed from reference 49.) The principal advantages of this method are that almost any amount of dust/unit time can be generated continuously and almost any material that can be powdered can be used. One of the major disadvantages of dry redispersion occurs because fine particles packed close together in the original powder will tend to agglomerate and resist separation. Sufficient energy must be supplied by the dispersing mechanism to cause agglomerates to be reduced into their component particles before being introduced into the test situation. This can be accomplished to a certain extent by proper dispersion equipment.

Almost all dry powders will consist of particles of different sizes so that aerosols made from them will be heterogeneous with respect to particle size, shape and specific gravity. The few methods which have been developed to produce homogeneous aerosols (*i.e.*, uniform in size, shape and specific gravity) are discussed separately. The methods described in this and the next section will ordinarily be expected to produce heterogeneous aerosols.

Aerosol generators supply a controlled amount of the material to be dispersed, usually combined with some entraining air to carry it to the main air stream for dilution. Material must be provided at a constant and reproducible rate and if the entraining air stream is also used to reduce agglomeration, it may have to be

[49] Air Pollution Handbook. Chap. 12. McGraw-Hill Book Co., N. Y. (1956).

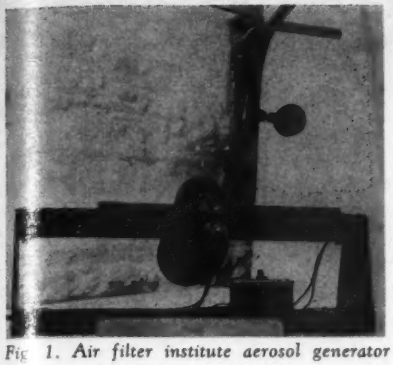


Fig. 1. Air filter institute aerosol generator

dried and supplied at fairly high velocity. Most feeders employ ejectors (or occasionally a stream of compressed air directed against the dust) by which dust is drawn up into a jet of compressed air and subjected to high impinging and shearing forces. It has been shown that higher air velocities in the jet will produce a lower percentage of agglomerates[1]. Agglomerates of several particles have a somewhat lower density than the constituent particles[2]. Dusts with a tendency to flocculate readily are frequently very difficult to disperse to their component parts. As a general axiom it might be anticipated that the smaller the ultimate particle size of the dust the less likely it will be to achieve this size by redispersion from a dry powder.

Some crystalline materials will shear on being subjected to a high velocity air jet, and the remaining pieces will possess a significant electrostatic charge tending to cause agglomeration in the subsequent aerosol. Many plastic powders will also acquire a high charge from the contact-separation mechanism. Whenever there is a possibility of unusual charge production on an aerosol by a generation process, this effect should be included in the analysis of results determined with the aerosol. The degree of dispersion obtained with a generator will depend upon the nature of the dust to be used and it is frequently impossible to get complete separation.

The simplest method for dispersion of dry powders consists of shaking the powder from a scoop into the moving air stream. Unless the particles are quite large this will not cause separation of agglomerates, and reproducibility of results will be low. Several types of solids feeders used in chemical processes[3] may be applied to provide uniform material flow, but these again may not guarantee adequate dispersion in the main air stream. An improvement on this method is available in the Deichmann Dust Shaker[4] which consists of a continuously-rapped screen into which dried dust is placed with the main air stream flowing underneath. In a similar device[5] the dust is placed on a sintered glass surface, agitated with a stream of air, and passed to an ejector.

Many generators use a compressed air ejector to remove dust from a moving trough, belt, turntable, or other constant feed device and cause it to be suspended in the expanding air stream. The ejector should be operated at pressures sufficient to attain high velocity (sonic if possible) at the point where the dust is introduced so that maximum particle separation will occur. A simple generator can be made with a vibratory feeder (Syntron) discharging directly into the pickup opening of an ejector. If the dust surface characteristics cause it to ball or clump in the vibratory feeder, another type of feeder may be required.

Table II includes the characteristics of many solid aerosol generators including those discussed below. Fig. 1 shows a device consisting of a horizontal trough on a rack and pinion drive. A rotating gear levels the dust in the trough and an ejector picks up and disperses the dust beyond the gear. A similar unit uses 4 grooves in a horizontal plate, with a dust-leveling gear and 4 ejectors. The feeder developed by the National Bureau of Standards, shown in Fig. 2, consists of a rotating gear with special teeth fed from a small hopper. An ejector removes the dust from the valleys between the teeth by means of a diagonal pickup pipe. These generators are used primarily for resuspending the NBS test dust (for air conditioning filter tests).

A turntable dust feeder used by the Harvard Air Cleaning Laboratory (for dust collector testing) is shown in Fig. 3. Material is fed from a vibratory feeder onto a slowly-rotating turntable; regulated to a constant width ribbon on the edge of the table by a scraper; and then passes to the pickup pipe of an ejector. This principle has been used by investigators of the American Society of Heating and Ventilating Engineers[6] and others[7].

A somewhat different feeding principle, used by the U. S. Bureau of Mines (for respirator testing)[8], is shown in Fig. 4. A mechanically-driven column of dust is fed to a spiralling air stream at the top of the column, and the entrained dust is fed to an ejector. Similar feeders have been developed by others[9,10].

Several investigators have used the centrifugal action of a small fan or blower to disperse free-flowing dusts into an air stream when it was desired to reduce the static electrification of the aerosol produced in high velocity air jets. In Fig. 5, a horizontal revolving disc of fine screen carries material from a vibratory feeder to an elutriating column. Dust is projected into the column by the action of a small blower located at the top, in conjunction with a compressed air manifold below the revolving screen, and the fine particles are dispersed into the main air stream from the blower outlet. A rapper on the side of the column prevents excessive

Fig. 2. National Bureau of Standards aerosol generator

[1] R. Dennis, et al. Air Cleaning Studies, Progress Rept. July 1953-June 30, 1954. USAEC Rept. NYO-4608, Harvard Univ., p. 17.

[2] W. J. Sheffy. Properties of Aerosol Agglomerates. Fourth Air Cleaning Seminar for AEC Personnel. Argonne Natl. Lab. (Nov. 1955).

[3] T. Olive. Solids Feeders. *Chem. Eng.* **59**, 163 (1952).

[4] W. B. Deichmann. A Dustshaker. *J. Ind. Hyg. Toxicol.* **26**, 334 (1944).

[5] L. S. Sonkin, et al. An Apparatus for Dispersing Finely Divided Dusts. *J. Ind. Hyg. Toxicol.* **28**, 273 (1946).

[6] F. B. Rowley and R. C. Jordan. Air Filter Performance as Affected by Kind of Dust, Rate of Dust Feed, and Air Velocity Through Filter. *Heating, Piping and Air Cond.*, **10**, 539 (1938).

[7] M. W. First, et al. Test of "Fiberfrax" Ceramic Fibers for High Temperature Gas Filtration. Proc. 48th Annual Meeting, APCA (1955).

[8] H. H. Schrenk. Testing and Design of Respiratory Protective Devices. U. S. Bur. Mines. Inf. Circ. 7086, (Sept. 1939).

[9] W. H. Smith and R. B. O'Brien. Special Test of Aerodyne Dust Collector. Third Air Cleaning Seminar for AEC Personnel. Los Alamos Scientific Lab. (Sept. 1953).

[10] B. M. Wright. A New Dust Feed Mechanism. *J. Sci. Inst.* **27**, 12 (1950).

dust accumulation[11]. A device, used by Research-Cottrell[12] for dispersing free-flowing materials by fan action, is shown in Fig. 6. It consists of a small squirrel-cage centrifugal blower mounted with its axis of revolution vertical and containing a horizontal circular plate on the inside of the impeller. A funnel placed above the center of the rotating plate feeds the dust down onto the plate and the action of centrifugal force and the inward-moving air cause the material to be carried out to the fan blades where it is dispersed. Material feed rate is controlled by the height of the funnel spout above the rotating plate.

Combinations of several of the above methods may be required for dusts difficult to disperse[13]. Continuous generation of flake iron powder was accomplished in this manner with the unit shown in Fig. 7. A special screw feeder made from a kitchen meat-grinder (with adjustable orifice plates located at the screw discharge) was used to feed powder onto a narrow moving belt. A rotating scraper blade controlled the width of the ribbon, and material was removed from the end of the belt by an ejector.

As has been suggested by some of the applications above and those given in Table II, it may be desirable to separate large agglomerates (or even large particles) by the use of intermediate size-selective collecting devices. Elutriating towers (or settling chambers) are frequently used to control the size of aerosols. Cyclones, impingement units, screens, or coarse filters may also be used to cut off aggregate sizes. Since all of these have some collection efficiency, they would be expected to reduce the overall dust loading to a certain extent, which might be a drawback.

Redispersions of Solids from Liquid Suspensions

Dispersion of a colloidal suspension (Table I, D) may form a solid aerosol by drying the resulting liquid droplet. The solid material must be finely ground and may require a wetting agent or mechanical means to keep it in suspension. A simple method for obtaining dispersion of fine liquid drops is to use a high pressure hydraulic (or pneumatic-hydraulic) spray nozzle. Any standard nebulizer, atomizer, nozzle, or paint or insecticide sprayer may be used but it is usually desirable to provide additional settling or jet impingement to eliminate coarse drops which may contain several particles (and which also tend to cause agglomeration). If the liquid is volatile (such as alcohol), it may be evaporated at ambient temperatures without any additional heating. Water is frequently used so the mist must be evaporated by passing through a heated drying tube.

Liquified gases are very convenient for spraying but their cost is usually prohibitive for large or extended experiments[14]. So-called aerosol spray *bombs* may be obtained which contain soluble salts, dyes, or insecticides in methyl chloride, freon, or other liquified gases.

The techniques above are suitable for low dust concentrations. For higher concentrations a Venturi atomizer[15] or the Aerojet-Venturi[16] may be supplied with the dust in a liquid suspension. A large scale nebulizer-ejector using 24 units in parallel has been developed[17] to supply salt crystals to large air volumes and could also be used for colloids.

Information on atomization[18] and a bibliography on spraying[19] is available.

Primary Formation of Solid Aerosols

Atmospheric dust is the most prevalent solid aerosol and is frequently used for test purposes. Many methods of measuring amounts of this aerosol by weight, stain, and count are in current use. Variations in this dust occur with time, weather, and industrial activity. A recent study of roughing filter performance[1] correlated particle size with local activity. Atmospheric dust concentrations and proportions of particles over 1 μ were established by sweeping the floor around the test unit at various rates. Filter efficiencies (by stain) showed a definite relationship to the proportion of particles greater than 1 μ and thereby to degree of local activity.

Attrition

Many dusts may be produced for laboratory investigations in the same

[11] L. Silverman, et al. Final Progress Rept. (Feb. 1, 1950). USAEC Rept., NYO-1527, Harvard Univ.

[12] Personal Communication.

[13] L. Silverman and I. L. Beauchamp. Open Hearth Stack Gas Cleaning Studies. Semi-Annual Rept. (SA-1) for Nov. 1, 1953 to Apr. 30, 1954. Harvard Univ.

[14] L. D. Goodhue. Insecticidal Aerosol Production. *Ind. Eng. Chem.* 34, 1456 (1942).

[15] E. W. Comings, et al. High Velocity Vaporizers. *Ind Eng. Chem.* 40, 74 (1948).

[16] R. M. G. Boucher. Sur le Functionnement de l Epurateur a Microbrouillards "Aerojet-Venturi." *Chaleur et Ind.* 33, 363 (1952).

[17] L. Dautrebande, et al. Essai de Prevention de la Silicose. Union Miniere du Haut-Katanga (1954).

[18] R. A. Castleman, Jr. The Mechanism of the Atomization of Liquids. Bu. Stan. *J. Rsch.* 6, 369 (1931).

[19] Pennsylvania State Univ., Dept. Eng. Research, Bibliography on Sprays. 2nd Ed. Pub. by Texas Company Refining Dept., Tech and Rsch. Div., New York (1953).

[1] See footnote 1, page 79.

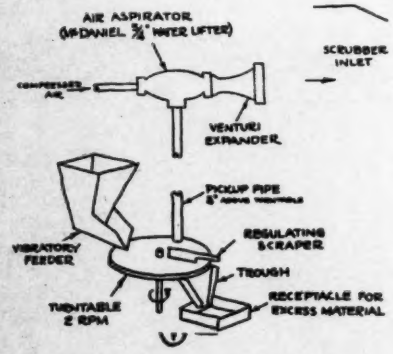


Fig. 3. Harvard turntable feeder-aerosol generator

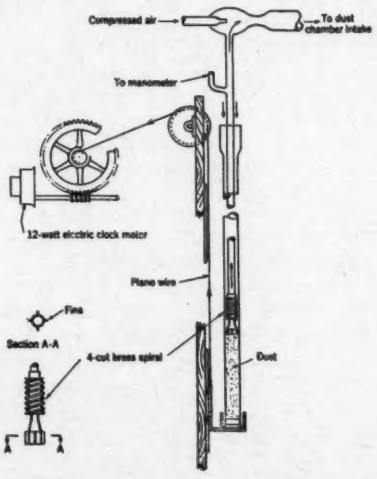


Fig. 4. Bureau of Mines aerosol generator (respirator tests)

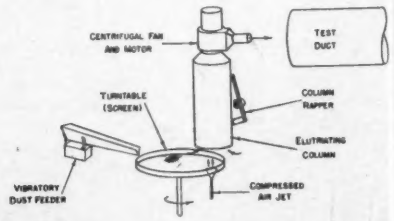


Fig. 5. Harvard elutriating column-gan aerosol generator

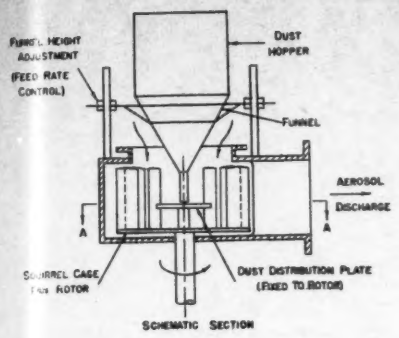


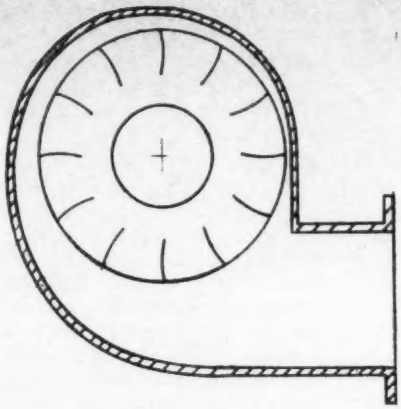
Fig. 6. Research-Cottrell fan aerosol generator

manner that they are produced naturally or industrially (*i.e.*, friction, impact, explosion, *etc.*). Small hammer or impact mills is used to crush or grind solid or agglomerated materials can be supplied with air to draw off the fines generated. Ball or rod mills may be used by passing air through them during operation. A rotating drum filled with charcoal (in conjunction with an elutriation column) produced controlled dust concentrations in the range of 250 mg./m.^{3} by varying drum speed. For large scale testing, energy mills[20] produce fine materials (near 1 μ) in quantities sufficient for 200 or more ft.^{3} of air/min. A number of companies produce laboratory or portable size-reduction equipment suitable for aerosol generation. Size reduction principles and applications have been given in a recent article[21].

Another attrition method[22] uses a steel wire brush revolving rapidly against an aluminum plate to produce a solid aerosol. An impaction plate followed this device to collect particles above 5 μ. Scraping, brushing, or grinding of many materials is possible as long as the material and moving surface are of widely different resistances to abrasion to prevent a 2-component aerosol with different properties.

Combustion

A large number of aerosols (Table I, E) are products of combustion of organic (usually) or inorganic (occasionally) substances. Aerosols produced by burning organic materials are frequently liquid droplets at room temperature containing tarry hydrocarbons and carbon.



SECTION A-A

Fig. 6a.

The incomplete combustion of a gas such as acetylene[23] or city gas[24] will produce the familiar carbon black aerosol which is a long chain of agglomerated carbon particles. Discrete carbon particles are very difficult to produce. Some liquids (benzene, *etc.*) will also produce carbon when incompletely burned.

Metals may be burned by many processes, such as: sparking in a high voltage arc; gas welding or flame cutting; or by combustion of metallo-organic compounds (lead tetraethyl, zinc ethyl, iron carbonyl). Lead tetraethyl may be volatilized into a natural gas stream and burned continuously to produce lead fume as done by the U. S. Bureau of Mines for respirator testing[25]. Iron pentacarbonyl vapor can be burned in a gas flame to yield spherical particles of iron oxide[26].

An increasing interest in uranium aerosols (from activities in the atomic energy industry) has led investigators to produce these by combustion of the metal. To achieve reproducible results, chips of uranium are fed slowly into an oxygen-argon gas mixture by a rotating wire screen turntable[27]. Uniform burning occurs and aerosols from 0.05 to 0.2 μ (geometric standard deviation 2.5 and 3.1 respectively) can be produced by varying mixture ratio and gas flow rate.

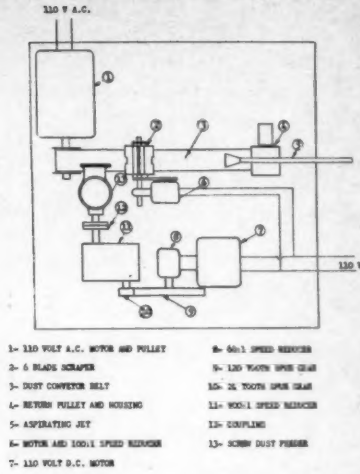

110 V A.C.
110 V.D.C.
1- 110 VOLT A.C. MOTOR AND PULLEY
2- 6 BLADE SCRAPER
3- DUST CONVEYOR BELT
4- RETURN PULLEY AND HOUSING
5- ASPIRATING JET
6- MOTOR AND 100:1 SPEED REDUCER
7- 110 VOLT D.C. MOTOR
8- 60:1 SPEED REDUCER
9- 120 TOOTH SPUR GEAR
10- 24 TOOTH SPUR GEAR
11- 900:1 SPEED REDUCER
12- COUPLING
13- SCREW DUST FEEDER

Fig. 7. Harvard combination feeder-aerosol generator

Metals that will sustain combustion spontaneously (such as magnesium) may be ignited to produce the metal oxide. Thermit and gas welding operations also produce metal fume aerosols. Inert-gas welding will produce a mixed aerosol containing the original metal with its oxides. Metal powders (lead, iron, *etc.*) have been injected into air-oxygen-acetylene flames to form aerosols for filtration studies[27]. The exothermic reaction of iron powder[13] in one case yielded irregular particles as opposed to spherical particles usually obtained. Most metal *smokes* from the above generators are in the size range 0.001 to 0.5 μ.

Combustion of organic materials (tobacco, rosin, *etc.*) produces a useful aerosol for test purposes. Tobacco smoke generators for static clouds[28], respirator testing[29], and continuous large scale testing[30] have been developed.

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[20] C. H. Wheeler Co., Phila. 32, Pa.

[21] J. C. Smith. Size Reduction. *Chem. Eng.* **59**, 151 (1952).

[22] J. J. Fitzgerald and C. G. Detwiler. Collection Efficiency of Air Cleaning and Air Sampling Filter Media. KAPL 1088. Knolls Atomic Power Laboratory, Schenectady, N. Y. (1954).

[22] S. H. Katz. Generator of Fine Smoke for Direct Impregnation of Filter Paper Used in Gas Masks. U. S. Pat. No. 2,528,522 (1950).

[24] A. S. G. Hill. A Photo-electric Smoke Penetrometer. *J. Sci. Inst.* **14**, 296 (1937).

[25] U. S. Bureau of Mines, Approval Schedule No. 21, Procedure for Testing Filter Type Dust, Fume, and Mist Respirators for Permissibility (1934).

[26] C. E. Billings, et al. Open Hearth Stack Gas Cleaning Studies, Semi-Annual Rept. (SA-3) for Nov. 1, 1954 to April 30, 1955. Harvard Univ.

[23] See footnote 13, page 80.

[27] P. LaTorre and L. Silverman. Collection Efficiencies of Filter Papers for Sampling Lead Fume. *Arch. Ind. Hyg. & Occ. Med.* **11**, 243 (1955).

[28] P. Drinker and T. Hatch. Industrial Dust. 2nd Ed. McGraw-Hill Book Co., N. Y. (1954).

[29] S. H. Katz, et al. Dust Respirators, Their Construction and Filtering Efficiency. U. S. Bur. Mines Tech. Paper No. 394 (1926).

[30] M. W. First, et al. Performance of Wet Cell Washers for Aerosols. *Ind. Eng. Chem.*, **43**, 1363 (1951).

[47] H. Glauberman and A. J. Breslin. The Generation of a Solid Sub-Micron Test Aerosol. Paper 43, AIHA Annual Meeting, Phila., Pa. (1956).

The particle size of the resulting aerosol is in the range 0.2 to 0.6 μ, with air concentrations about 100 mg./m.^{3}.

Molten metals or plastics from spray metallizing guns disperse into stable aerosols. Zinc, lead, iron, brass, and aluminum have been used to produce aerosols about 3 μ in size. Plastics and inorganic (ceramic) frits may also be aerosolized by the flame spraying process. Equipment is available commercially and the techniques have been described[31,32].

Condensation

Aerosols can be generated from many materials by heating to volatilization and then allowing the vapors to condense (Table I, C). Waxes or resins with reasonably low vaporization temperatures can be made into spherical or crystalline aerosols by this simple method. Several of the metal aerosols discussed in the previous section can be produced from a molten bath. In many cases the metals will oxidize (at the necessary high temperatures) and produce changes in the aerosol surface characteristics. Lead and zinc will condense as the oxide unless prepared in an inert atmosphere (nitrogen, *etc.*) in which case spheroids of the metal are produced[33]. Atomized mercury and tetramethyl lead have been treated with a mercury vapor lamp[34] to produce condensation aerosols.

Evaporation of Liquid Solutions

In the section above on wet redispersion of previously ground powders, it was indicated that there are many methods for producing a liquid spray. If the substance to be generated (salt, resin, *etc.*) is dissolved in a reasonably volatile solvent, the solution can be sprayed and dried to form a solid aerosol directly. The aerosol size will depend on the original liquid droplet size and the concentration of the substance in the solution. The final size of the solid will be equal to the original droplet diam. multiplied by the cube root of the concentration. The number of droplets produced/unit of time will be equal to the liquid volume feed rate divided by the average vol./droplet produced. If the concentration from a single nozzle is too low, a multiple nozzle system may be required. An elutriation chamber or an impingement device should be used after the spray to remove coarse droplets, as indicated previously.

Sodium chloride (and other salts) in water or methylene blue dye in alcohol have been used in this way[35,36]. If the drying is rapid (as in the case of alcohol solvent) spheres frequently result. When air-dried at room temperature salts frequently yield cubic particles. By passing the spray droplets through a heated column and raising the material above the melting point, the resulting aerosol will fuse and become spherical. Sodium chloride, copper sulfate, and uranium oxide have been produced as spherical aerosols by this method. Hydration of some salts will gradually cause them to resume crystalline shapes (in about 24 hr. in the case of copper sulfate).

In the *spinning top* aerosol generator a thin stream of liquid is allowed to run onto the center of a rapidly rotating solid disc. The liquid is discharged from the edge of the disc as fine droplets[37] in 2 distinct categories; a major particle and a smaller satellite. Extraction of the satellite[38] leaves a uniform liquid spray and if it contains a soluble salt, dye, or resin in solution, can give a solid aerosol upon evaporation. In another case the extracted satellites were used by impinging the larger drops to produce a methylene blue aerosol of small size[39].

Homogeneous Aerosols

Research in the fundamental laws governing aerosol behavior under the action of various forces is facilitated by the use of aerosols with constant and controllable size, shape, and specific gravity. This not only simplifies the number of variables to be considered but makes the mathematical expression of the relationships clearer (the variations of parameter D indicating 1 particle size) is considerably more apparent than variation of the complex expotential form required for a particle size distribution). Unfortunately it is difficult at present to generate homogeneous solid aerosols, particularly in high enough concentrations to provide usable information in large scale tests.

For most applied research and engineering investigations the techniques described above for heterogeneous aerosol production are more widely used, particularly when it is desired to simulate naturally or industrially-occurring aerosols.

Homogeneous liquid aerosols are usually produced by 1 of 2 methods, both of which could be used for solids in liquid suspensions or for soluble salts, resins, *etc.* In the LaMer generator[40] a liquid is vaporized under close control, allowed to condense on nuclei, and then caused to grow to the desired size in saturated vapor. This is adaptable to solid substances which can be caused to sublime and condense on nuclei. Uniform aerosols have been produced from oleic and stearic acid, triphenyl and trichresyl phosphate, rosin, menthol, ammonium chloride, lubricating oil and Aroclor. At present micro-crystalline waxes and volatile dyes are being investigated by LaMer. This generator yields homogeneous particles in the range 0.2 to 10.0 μ. Concentrations are in the range 1 to 10 mg./l. from the generator in 1 to 4 l./min. The aerosol must be diluted 10 or 100 times to prevent agglomeration.

The spinning top (rotating disc) device described in a previous section generates a homogeneous droplet with a smaller (less homogeneous) satellite. If these contain a salt or resin in solution then evaporation of the solvent will leave a homogeneous solid aerosol. Drop sizes range from 15 μ to several hundred and the solid particle size is proportional to the cube root of the concentration.

Dry or wet dispersal of uniform-sized particles may be used for homogeneous aerosols if care is taken in generation to remove or reduce all agglomerates. Colloidal suspensions of latex and resin particles have been made for calibration purposes in electron microscopy, *etc.*, but are too expensive for general use. In order to use wet dispersal methods the solution must be quite dilute so that

[31] H. S. Ingham and A. P. Shepard. The Metco Metallizing Handbook. Metallizing Engineering Co., Long Island City, N. Y. 5th Ed. (1951).

[32] H. I. Miller, et al. Health Hazards in Metal Spraying. *J. Ind. Hyg. Toxicol.*, **20**, 380 (1938).

[33] R. G. Keenan and L. T. Fairhall. The Absolute Efficiency of the Impinger and the Electrostatic Precipitator in the Sampling of Air Containing Metallic Lead Fume. *J. Ind. Hyg. Toxicol.* **26**, 241 (1944).

[34] P. Nagel, et al. Condensation Aerosols: Mercury Fog and Lead Oxide Smoke. *Kolloid Z.* **107**, 194 (1944).

[35] W. H. Walton. The Methylene Blue Particulate Test for Respirator Containers. Porton Spec. 1151. Chem. Def. Cent., Porton, Eng. (1940).

[36] W. H. Walton. The Sodium Flame Particulate Test for Respirator Canisters. Portion Spec. 1206. Chem. Def. Cent., Porton, Eng. (1941).

[37] W. H. Walton and W. C. Prewett. Production of Sprays and Mists of Uniform Drop Size by Means of Spinning Disc Type Sprayers. *Proc. Phys. Soc.* (London), **628**, 341 (1949).

[38] K. R. May. An Improved Spinning-Top Homogeneous Spray Apparatus. *J. Appl. Phys.* **20**, 932 (1949).

[39] A. T. Rossano, Jr. and L. Silverman, Electrostatic Mechanisms in Fiber Filtration of Aerosols. USAEC Rept. NYO 1594, Harvard Univ. (1955).

[40] V. K. LaMer. The Preparation, Collection and Measurement of Aerosols. Proc. First Natl. Air Poll. Sym., Stanford Rsch. Inst., (1949); and D. Sinclair. Handbook of Aerosols. Chap. 6, USAEC. Wash., D. C. (1950).

each drop will contain only 1 particle, which limits the concentrations that can be used. Dry dispersion subjects particles to static electrification and this may prevent reduction of agglomerates.

Many bacteria are fairly uniform in size and in some cases are spherical in shape. These can be sprayed from solutions by nebulizers and other techniques[41]. Pollens and spores are also used occasionally. In either case the amounts that can be generated are quite small and offer complications in laboratory handling and sanitation. A rotating cylindrical screen with mechanical impact used to produce discrete pollen particles is shown in Fig. 8.

The use of sonic energy to redisperse dry powders is mentioned in Table II. A recent publication[45] has suggested the use of ultra-sonic energy for the production of homogeneous aerosols. Preliminary investigation of the role of physical factors such as vapor pressure, surface tension, viscosity, and generator frequency in controlling size distribution of the aerosol has been accomplished and the results are promising.

The major limitation of all of the above methods is that only small or limited amounts of materials can be generated which limits the maximum concentration available. For large air volumes the concentration would be quite low.

Comparison of Redispersion and Primary Formation Methods

There are definite limitations in the generation of aerosols from previously-formed powders and in the direct or primary formation methods which have been discussed. In many cases it is desired to simulate aerosols found industrially (or naturally) which are a product of some primary formation process. When this is done in the laboratory by redispersing a dry powder, the particle size may be quite different.

This section compares some aerosol particle sizes produced by 2 generation methods from:

(1) a chemical operation
(2) vaporization of a metal, and
(3) combustion of pulverized coal.

Calcium Carbonate

Precipitated pure calcium carbonate from a commercial supplier was redispersed by the turntable feeder and air ejector shown in Fig. 3. This was used

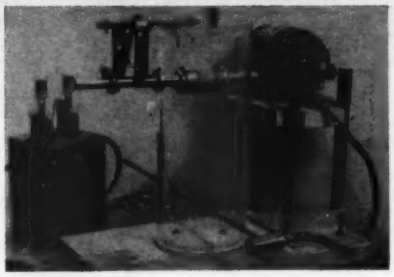

Fig. 8. Harvard rotating screen pollen generator

to test high efficiency dust collecting equipment[42] and particle size samples were taken from the air stream on Millipore filters. The mass median diam. of this test dust was 2.6 μ (geom. std. dev. 2.0). Electron microscope samples of the individual particles taken during manufacture of the precipitate indicate an ultimate particle size of 0.03 or 0.04 μ[43]. It is apparent that the powder did not become completely deagglomerated in passing through the single ejector stage. This calcium carbonate is made commercially by combining pure solutions of calcium chloride and sodium carbonate to yield an ultra-fine precipitate in a sodium chloride solution. Since this method yields the precipitate in discrete particles in a liquid, it should be possible to produce the calcium carbonate as an aerosol by spray dispersion and drying of the liquid suspension. In order to avoid producing crystals of sodium chloride the salt in the liquid would have to be removed. Although this procedure was not used to produce a calcium carbonate aerosol, it was suggested for this study. The primary drawback was felt to be one of limitation in the amount of solid that could be made. Concentrations in the range of 100 to 500 mg./m.^{3} were required in large air volumes.

Vaporized Silica

In the production of silicon metal or ferro-silicon alloy, crushed quartz is melted at high temperature in an electric furnace. Some fume escapes around the electrodes to become airborne as a vaporized silica aerosol (amorphous silicon dioxide). The particle size of this material has been found to be about 0.3 μ[44] and the electron microscope shows that these are agglomerates of many smaller particles[45]. As in the case of other metal fumes (magnesium or zinc) the formation of the oxide apparently causes a rapid agglomeration. This aerosol is collected in some cases and sold as a filler or extender. The commercial product was used to test a fabric dust filter. Material from the shipping bag was found to be quite coarse and gritty. This was aerosolized with the generator in Fig. 3, recovered from the dust collector, and regenerated about 10 times. At the end of this period the material showed a significant decrease in apparent bulk density and felt very light and fluffy. The several successive aerations produced a powder very similar to that collected from the actual electric furnace operation. It was estimated that the particle size of this reworked powder was about 0.4 μ. Resistance of the filter cake to air flow at the end of the period was significantly above that at the start, indicating a decrease in particle size in the cake (using the Kozeny specific resistance concept).

Fly Ash

Fly ash from the Cottrell precipitators of public utility plants has been frequently used as an aerosol. (The National Bureau of Standards filter test dust is obtained in this way.) The ash is resuspended with air ejectors (and dust feeders) described in a previous section, where it was indicated that the air velocity in the ejector was of some importance in breaking up agglomerates. In order to further investigate the relationship between the normal production of a fly ash aerosol and the laboratory resuspension, samples of the resuspended aerosol were compared with the aerosol from the direct combustion of pulverized coal.

A particle size analysis of the NBS test aerosol (from a Millipore filter sample) indicated a mass median diam. of 10.0 μ (geom. std. dev. 2.73) when resuspended with the generator shown in Fig. 3.

Pulverized coal was burned at a uniform rate by dropping it from a vibratory feeder into a carbon arc located at the base of a chimney[46]. The resulting fly ash aerosol appeared to be the same as that from resuspension of collected material. The particle size of this aerosol was approximately 8.0 μ (geom. std. dev. 3.5). Efficiency of a centrifugal collector tested with both aerosols was slightly lower with the freshly generated fly ash, which was believed to be due to a lower percentage of agglomerates.

[41] T. Rosebury. Experimental Air-borne Infection. Microbiological Monographs. Williams and Wilkins Co., Baltimore, Md. (1947).

[42] D. A. Fraser. Ultrasonically Produced Aerosols for Animal Experiments. Paper 32. AIHA Annual Meeting, Phila. Pa. (1956).

[43] C. E. Billings, et al. Laboratory Performance of Fabric Dust and Fume Collectors. USAEC Rept. NYO 1590, Harvard Univ. (1954).

[44] Purecal in GR-S. Wyandotte Chemicals Corp., Wyandotte, Mich.

[45] L. Silverman and R. A. Davidson. Electric Furnace Ferrosilicon Fume Collection. *AIMME Trans.* **203**, 1327 (1955).

[46] M. O. Sem and F. C. Collin. Fume Problems in Electric Smelting and Contributions to Their Solution. *Proc. 48th Annual Meeting*, APCA (1955).

[47] Harvard Univ. Air Cleaning Laboratory, Unpublished data (1956).

A Study of the Effluents From Back-Yard Incinerators*

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Those who have studied the smog problem in the Los Angeles area are generally agreed that back-yard incinerators are important contributors to the over-all air-pollution problem. However, they cannot agree as to the exact extent to which this type of refuse disposal contributes to smog. Certainly the burning of a heterogeneous mass such as household trash under a variety of combustion conditions can be expected to emit an extremely complex mixture of products, many of which are highly reactive. It is reasonable to assume, therefore, that compounds originating in incinerator effluents are capable of entering into smog-forming reactions.

The results of studies by the Los Angeles County Air Pollution Control District and Stanford Research Institute (1) have indicated that a substantial portion of the effluent from back-yard incinerators can be classed as organic materials. In order to obtain more information as to the nature of these effluents, the Air Pollution Foundation sponsored a 1 yr. research program at Battelle Memorial Institute. This paper describes the work which had as its objective the determination of the amounts of various materials emitted from a typical back-yard incinerator, with special emphasis on the analysis of the organic fraction of these effluents.

Equipment and Procedure

Operation of the Incinerator

An incinerator, typical of those used in the back yards of Los Angeles, was built and set up in the laboratory for this study. Auxiliary equipment for the measurement of burning conditions and the collection of effluent from the incinerator was installed in a manner that would not interfere with the normal operating characteristics or draft of the incinerator. A mixture of paper, grass clippings, hedge trimmings, and similar materials commonly burned in the back-yard incinerators in the Los Angeles area was used for the incinerator tests.

Fig. 1 shows a sectional view of the incinerator. Constructed of firebrick, it had a capacity of 6 ft.^{3} between the grate and the door sill. Air for combustion of the charge entered under the grate through a draft tube that housed the sensing element of an anemometer. The temperature of the effluent gases was measured in the short stack at the top of the incinerator, and samples of the gases were withdrawn near the top of the stack.

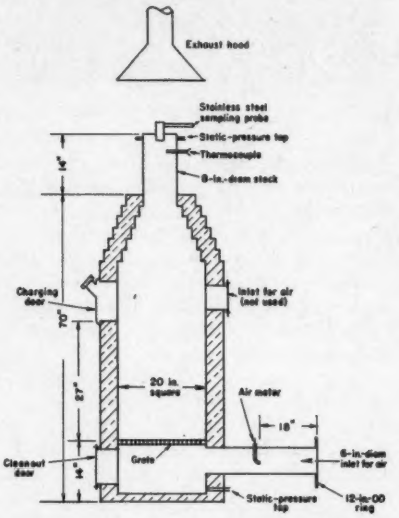

Fig. 1. Sectional view of the typical backyard incinerator built for use in this study

Fig. 2 is a photograph of the incinerator and auxiliary equipment used in this study. The incinerator is in the foreground, with the air-inlet duct and air meter to the bottom left. One version of the sampling train is at the right of the incinerator and the instruments for recording the draft and the temperature of the effluent are in the right background.

Table I shows the composition of the *standard* charge mixtures burned in the incinerator. During the summer months a greater variety of green vegetation became available locally and mixture *A* was replaced by mixture *B* which was more representative of the refuse burned in the Los Angeles area. The most significant phase of the study was conducted with mixture *B*.

Table II shows the test charge and combustion data for each of the sampling runs. Each charge was ignited at the top and burning was usually intense at the start of a run. As the paper and easily-ignited materials were consumed, the charge smouldered and at times required reignition. The effluent contained visible smoke except for the short periods of rapid burning. Although care was taken in loading each charge to obtain the same distribution of paper and vegetation throughout the charge, and the same degree of packing, the results show a wide range in burning time, air-fuel ratio, and temperature of the effluent.

In order to relate the volume of effluent produced to the quantity of charge burned in the incinerator, the assumption was made that the volume of effluent was the same as the volume of air required for combustion, when volumes were corrected to normal temperature and pressure. The flow rate of air for combustion was measured continuously with an anemometer located in the intake duct as shown in Fig. 1. The total volume of air supplied in each sampling run was divided by the weight of charge to obtain an average air-fuel ratio which was then used as the basis for expressing the analytical results in terms of pounds of material in the effluent/ton of charge burned. Average air-fuel ratios were also used for the results of *grab* samples, as the specific air-fuel ratio during different periods of a test run was not known.

Development of Equipment and Sampling Procedure

During the first 5 sampling runs efforts were made to collect samples of normally gaseous and normally liquid materials from the effluent in a conventional laboratory freeze-out train whose traps were connected in series and maintained at successively lower temperatures with ice, dry ice, and liquid nitrogen. The capacity for flow in these traps was only a few l./min., and difficulties occurred repeatedly from the accumulation of ice in the small-sized glass tubing.

In the analytical phase of the work it was found that relatively large samples of the total would be required for developing a chemical separation technique. Therefore, the entire sampling train was rebuilt to permit sampling at peak rates of flow up to 5 cfm. Trials with the larger sampling train during runs 6 and 7 indicated the need for further modifications which were then made. During runs 8 through 10 the sampling train was operated at an average flow rate of about 3 cfm. with some difficulty from

(1) Air Pollution Foundation, Report No. 2, "Combustion and Smog."

* Presented at the 49th Annual Meeting of APCA held at Buffalo, N. Y., May 20-24, 1956.

plugging of the filters. For this reason, additional filter capacity was installed for runs 11 through 14. In each series of runs, 8 through 10, and 11 through 14, a sample of condensate was composited in the freeze-out train.

Fig. 3 shows a view of the sampling train in its final form as it was used in runs 11 through 14. The containers for the cooling baths were lowered for this photograph to expose the freeze-out traps.

In order to collect a representative quantity of condensate from a series of runs, a sample of effluent was drawn through the train at a rate proportional at all time to the rate at which the effluent was discharged from the incinerator. This was done by manually regulating the sample flow rate in accordance with the fluctuations indicated by the anemometer. The diameter of the sampling probe was such that isokinetic sampling conditions were provided. In runs 11 through 14, 888 ft.^{3} of effluent, which represented 5.9% of the total effluent, was passed through the sampling train at an average flow rate of 2.5 ft.^{3}/min.

Fig. 4 is a diagram of the sampling train. With the exception of the stainless steel probe, the train was fabricated from Pyrex glass. The minimum size of glass tubing used in the traps and connecting tubing was 15 mm.

TABLE I

Composition of "Standard" Charge Mixtures

	Charge Mixture "A" % by Wt.	Charge Mixture "B" % by Wt.
Newspapers	25	25
Cardboard	5	5
Rags	1	1
Grass Clippings	0	10
Fresh flower cuttings	55	17
Tree branch trimmings	0	20
Hedge trimmings	0	20
Scraps of wood	14	2

As much water vapor as possible was removed from the gas sample in the liquid state in ice traps 1 and 2 by providing reserve cooling capacity to maintain exit gas temperature close to 0 C. A short column of Raschig rings was included in the second trap. Traps of larger bore were found to be necessary in the next section of the train to avoid plugging. It was also believed that gradual cooling would minimize the formation of small crystals of ice which were difficult to filter. Accordingly, trap 3 was maintained at an intermediate temperature of about -29° C, which gave exit gas temperatures of from -10 to -20° C. A glass rod in the form of a helix was used in each U trap.

The second U trap, shown as No. 4, was maintained at dry-ice temperature. It contained a filter which had sufficient area to accumulate the ice and tarry material from an entire sampling run. The filter consisted of a 10-in. length of perforated glass tubing, 1 in. in diameter, which was wrapped with a ¼ in. layer of Pyrex glass wool. The temperature of the gas sample at the exit of this trap was about -60°C.

The final trap in the train was a coil trap having a filter section containing Pyrex glass wool. Exit gas temperatures in this trap were from -65 to -70°C. A reheat coil was used to obtain gas at room temperature for measurement in a rotameter. The maximum pressure loss in the train was 3 in. of mercury.

During the entire period of time that a series of runs was in progress, the last coil trap, 5, was held at the temperature of dry ice. After each sampling run in the series the train was sealed, and trap 4 was brought to room temperature before replacing the glass wool filter. The filter with its condensate was stored in a refrigerated container and became a part of the composite sample at the end of the series of runs.

In addition to the use of a freeze-out train for sampling and concentrating the materials in the effluent, direct determinations were made of certain materials in the raw effluent. In these instances, grab samples of the effluent were drawn into evacuated flasks or directly into the instrument. These methods, together with the analytical procedures, are discussed in the following section.

The Analysis of Incinerator Effluents

In studying the types of materials that are emitted from a back-yard incinerator, the effluents were considered generally to be either normally gaseous or normally liquid, although considerable overlap of compounds between the 2 categories could be expected.

Fig. 2. The backyard incinerator and testing equipment

TABLE II

Summary of Test Conditions and Data For The Sampling Runs

Run No.	Date 1955	Wt. of Charge, lb.	Standard Charge Mixture[a]	Average Air-Fuel Ratio, cu. m./lb.	Burning Time min.	Temperature of effluent Average F	Maximum F
1	1-25	17.4	A	—		180	900
2	2-3	18.2	A	—	100	230	1200
3	2-17	20	A	14.7	174	170	825
4	2-19	38.2	A	—	—	—	—
5	2-21	30	A	6.8	168	230	625
6	4-6	20	A	—	—	—	900
7	4-28	20	A	—	—	—	950
8	4-29	20	A	8.7	125	180	670
9	4-29	20	A	5.6	79	230	475
10	5-3	20	A	6.7	93	220	450
11	5-24	20	B	6.7	131	190	810
12	5-25	20	B	6.6	113	220	620
13	5-26	20	B	3.1	48	380	880
14	5-26	20	B	5.0	64	290	860
15	7-5	20	B	10.4	140	210	700
16	7-7	20	B	7.2	210	190	570
17	8-4	20	B	7.4	98	330	1110
18	8-23	20		6.5	103	410	1300

[a] A and B are the "standard" charge mixtures shown in Table I.

Analysis of Gaseous Products by Infrared Spectral Analysis

Materials Collected in Freeze-out Traps

A limited amount of information on the composition of incinerator gases was obtained by use of the infrared spectrometer for analysis of gases collected by condensation in a liquid-nitrogen-cooled trap during run 2. Other than CO_2, the materials identified were methyl alcohol, acetaldehyde, and carbonyl sulfide. Rough values for concentrations could be estimated only for CO_2 1%), methanol (20 ppm.), and acetaldehyde (90 ppm.). These results could be considered only as first approximations.

The major effort in the over-all sampling program was expended in gathering large samples of materials condensed from the incinerator effluent by means of a large-scale freeze-out train. In order to have available for analysis as much condensate as possible, the condensates from several runs were composited in the freeze-out train. Because of the high rate of sampling (about 3 cfm.), the use of a final liquid-nitrogen trap for the total condensation of some of the gases was not practical. Since the final traps in all variations of the freeze-out train were cooled with dry ice plus some solvent, some of the normally gaseous constituents were condensed out in these traps. After completion of a series of sampling runs, the sampling train was connected in series to a small Shepherd trap followed by a vacuum pump. The trap was cooled by immersion in liquid nitrogen, and was maintained at reduced pressure to prevent the condensation of oxygen from residual air in the traps. The sampling train was then brought slowly to room temperature, and held there for about an hour while the vapors evolved were condensed in the cooled traps.

The gaseous materials evolved from the composited condensate from runs 8-10 and runs 11-14 were studied separately by infrared absorption techniques. These analyses provided only qualitative data as to the types of normally gaseous materials which could be condensed in a dry-ice-cooled freeze-out trap. Gases from these runs showed the presence of acetone, methanol, benzene, and an ester, probably ethyl, propyl, or n-butyl acetate.

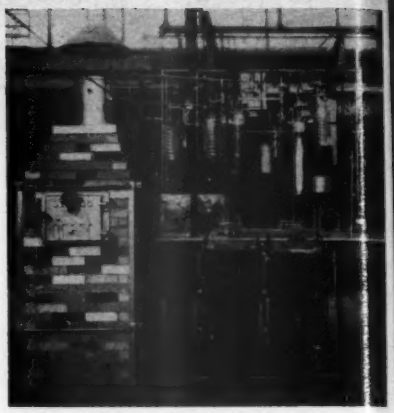

Fig. 3. The train of freeze-out traps used for collection of condensate

Analysis of Raw Incinerator Gases.

The most fruitful use to which infrared analytical techniques were put in this work was in the analysis of raw incinerator gases which had been collected in evacuated bulbs. This technique was used in runs 16 and 17.

In collecting samples for these analyses, evacuated gas bulbs of various capacities were allowed to fill with incinerator gases. A sufficient number of bulbs were used to permit the *grabbing* of samples several times during the incinerator runs. The contents of these bulbs were then drawn into an evacuated 1-m. infrared cell. The pressure in the cell was adjusted in accordance with the volume of the sample. In general, the concentrations of specific materials which could be identified were estimated from the intensity of the absorption bands and the pressure of the cell.

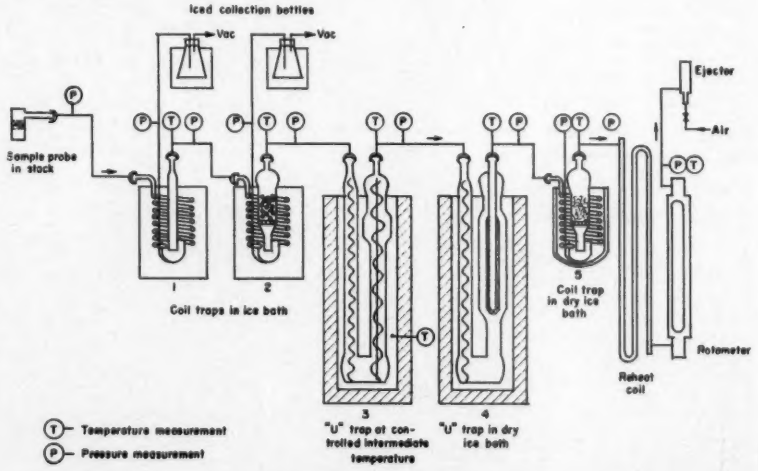

Fig. 4. Diagram of freeze-out train

Table III shows the results of the infrared analysis of samples of raw incinerator gas collected in evacuated sampling bulbs during run 17.

Most of the data were obtained on the spectra of the raw gases as expanded from each of the bulbs into the 1-m cell. To confirm the presence of materials identified in this way, and to search for additional compounds, the gases from a series of bulbs were composited by condensing in a liquid-nitrogen-cooled trap, and then re-expanded into the cell in a more concentrated form. Because of losses in the condensing operation, the amounts of all new materials which were identified after this treatment (α—olefin and carbonyl sulfide) were undoubtedly less than in the raw gas sample. Some of the composite samples were passed over Ascarite to remove CO_2 which masks a portion of the spectrum. Only in this way was it possible to identify benzene, which otherwise would have been undetected. The concentration shown is much less than that which would be expected in the raw gases because of excessive losses in this procedure. It was not possible to estimate the extent of benzene losses; however, it was noted that approximately $\frac{2}{3}$ of the acetylene was lost in the condensing operation while another 1/6 of it was lost in the Ascarite treatment. One would expect a much more efficient recovery of benzene in the condensation operation because of its lower vapor pressure. It is reasonable to assume that the benzene concentration in the raw gases is probably in excess of 0.003%. The recovery of methanol in the freeze-out trap was satisfactory, since the figures based on the liquid phase agreed well with those found in the gas phase.

Fig. 5 and 6 are infrared spectra from 2 typical samples of this type. Fig. 5 is the spectrum of the first bulb sample taken during run 17, and Fig. 6 is the spectrum of the composite gases taken in the first group of samples after condensation in a liquid-nitrogen-cooled trap. It will be noted that the peaks for condensable materials are intensified after condensing and compositing, but many of the extremely light materials such as methane and carbon monoxide were lost in this treatment. Because of the extreme complexity of the effluent mixture, the estimation of total *hydrocarbons* on the basis of the C-H stretching band intensity was not felt justified.

TABLE III

Results of Infrared Analysis of Raw Incinerator Effluent Sampled in Evacuated Flasks During Run 17 and Examined in a 1-M Cell

Material Identified	Concentration mg./m.³ of effluent	Emission, lb./ton of charge burned
Methanol	290-720	9-23
Ethylene	250-1870	8-61
Acetone	<260	<8
Methane	1,500-4,600	50-150
Carbon monoxide	16,000-30,000	530-1000
Acetylene	<120-1850	<4-6
φ—olefin (as propylene)	<190	<6
Carbonyl sulfide[a]	>80	>3
Benzene[a,b]	>105	>3

(a) The concentrations of these materials could be estimated only after their concentration in a liquid nitrogen cooled trap. Results shown are probably minimum values.
(b) Because CO_2 masked the spectrum, benzene could not be estimated in the raw gases. Determination was made after passing the gases through asarite. In this treatment, considerable loss of benzene could be expected.

Chemical Analysis of the Normally Liquid Portions of the Effluents

Once a considerable amount of material had been accumulated in the freeze-out train by combining condensates from a group of runs, the normally gaseous materials were removed by volatilizing at room temperature into a liquid-nitrogen-cooled trap for subsequent infrared analysis. This procedure was described earlier. The resulting condensate along with water and ether trap washings was examined by an organic chemist.

The first attempt at studying these normally liquid organic materials was made on the combined condensates from runs 4 and 5 which amounted to 255 ml. of collected material from 285 ft.³ of incinerator effluent. Most of the condensate consisted of water, and only about 2 ml. of the organic fraction could be recovered in extractable material. During the course of the analysis certain of the components gave positive tests for aldehydes and ketones and for phenols. The 4 extracts from the different treatments were each examined by infrared spectroscopy. The spectra of these samples showed them to be extremely complex mixtures; however, there was evidence for one or more acids, alcohols, and phenols. It was obvious from this preliminary work that larger samples of condensate and an improved separation technique would be required.

An improved version of the separation scheme was tried on the combined total condensates from runs 8-10. The starting material consisted of 960 ml. of aqueous condensate and 890 ml. of ether washings. This amount of condensate was based on 833 ft.³ of incinerator effluent passed through the system. The organic portions of the condensate were extracted as basic, neutral, phenol, and acid fractions which measured 1.5, 19.5, 2.5, and 6.0 ml., respectively. Attempts were made to distill each of these fractions at reduced pressure, but considerable decomposition was evident. Except in the case of the neutral fraction which distilled more readily, recoveries were less than 50%. The infrared spectra of the distilled fractions led to the identification of acetic acid, and gave some evidence for the presence of guaiacol, but generally the group separations were not sufficiently clean-cut to permit isolation of specific groups of compounds. The neutral fraction contained considerable amounts of phenols, although the alkaline extraction procedure was intended to remove them. The odor of the basic fraction would lead one to suspect the presence of pyridine or substituted pyridines; however, the infrared spectrum of

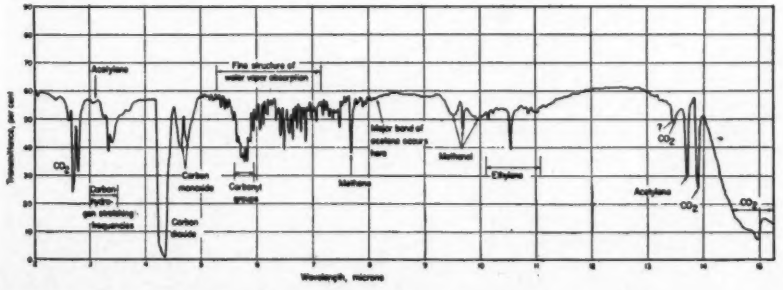

Fig. 5. Infrared absorption spectrum of raw gases from backyard incinerator run no. 17 (7 to 9 min. after starting test)

this fraction showed no real evidence for this type of material. On the basis of this experience it was believed that steam distillation instead of continuous solvent extraction would prevent some of the decomposition of fractions on distilling, and permit cleaner separation of the fractions.

Fig. 7 shows the improved scheme of analysis used to treat the 1245 ml. of liquid condensate plus washings collected from 888 ft.^{3} of incinerator effluent in runs 11-14. This scheme gave measurable amounts of several classes of organic compounds separated from the total condensate. The ether washings were concentrated by distilling to a head temperature of 47 C and discarding the distillates. The residue was combined with the freeze-out trap condensate and the water washings. After acidifying the mixture with HCl to pH < 2, it was distilled until only one phase appeared in the distillate. The distillate was then continuously extracted with ether, and the extract was concentrated to remove the solvent. The solvent-free material weighed 20.5 gm. and was submitted for chemical analysis of titratable acidity (3.8 gm. as acetic acid), carbonyl (0.1 gm. as formaldehyde), and phenols (9.0 gm. as phenol by ultraviolet spectroscopy).

Table IV lists the results of these analyses as rates of emission based on the volume of gases evolved and on the amount of material burned. The value for aldehydes shown cannot be compared with any other figures shown in this paper for these compounds because of the loss of low-boiling materials during the sampling and subsequent chemical treatments. As would be expected from the combination distillation-combustion process using wood and vegetable products, the amount of phenols in the incinerator effluent is substantial.

During the analysis of the condensates from runs 11-14, the basic fraction was accidentally lost and could not be analyzed further. This fraction was an evil-smelling mixture reminiscent of nitrogenous and amine substances, but could not be identified.

TABLE IV

Analysis of Normally Liquid Fraction of Condensate from Runs 11-14

Materials Analyzed For	mg./m.^{3} of Effluent	Lb./Ton of Charge Burned
Aldehydes and ketones as HCHO by titration	3.8	0.09
Titratable acidity as acetic acid	151	3.6
Phenols as phenol by ultraviolet	356	8.4

Analysis of Effluent Materials Collected in Evacuated Flasks

A method of analysis similar to that employed by the Stanford Research Institue[2] to study incinerator effluents was used to make direct determinations of ammonia, oxides of nitrogen, and aldehydes in runs 15-18. In these tests, reagents were placed in separate 5-gal. bottles which were then evacuated to a pressure of about 5 in. Hg. The reagents consisted of 2% H_2SO_4 for ammonia, 1% KOH and 1% H_2O_2 for oxides of nitrogen, and 1% $NaHSO_3$ for aldehydes. After obtaining the samples of incinerator effluent, the bottles were rolled and shaken intermittently for about 1 hr., after which each reagent was removed and submitted for analysis.

Table V gives the complete results of samples taken by this method during runs 15-18. The values shown for ammonia, ranging from 28 to 84 mg./m.^{3} of effluent, are in reasonable agreement with the results shown in the Stanford report; 3 and 100 mg./m.^{3} for paper and grass clippings, respectively. The higher concentrations were found during the later stages of burning, after the paper in the mixture of refuse had burned away.

The values obtained in these tests for the concentration of aldehydes, ranging from 182 to 1960 mg./m.^{3} of gases, are considerably higher than the Stanford results, which showed values of 60 mg./m.^{3} for paper and 125 mg./m.^{3} for grass clippings. In addition, the present work indicates that the production of aldehydes is greater while the paper and easily combustible portions are burning at an early stage of the test, as shown by the results of the analyses for aldehydes of successive samples during run 18. This could be the result of progressive changes in the type of material remaining in the incinerator and changes in burning rates and temperatures in the incinerator.

The extremely low concentrations of oxides of nitrogen found in this work are at variance with results of the earlier work done by the Stanford Research Institute and others which showed the concentration of oxides to be 13 or more mg./m.^{3} No explanation can be given for this difference.

Discussion of Results

Table VI summarizes the analytical data obtained during the course of this investigation, expressed as ranges of concentrations of the materials found in the effluent gases. The use of single average values for the concentrations does not appear to be justified because of the large variations in the values obtained at different periods of a single test occasioned by changing burning conditions,

[2]Third Interim Report on the Smog Problems in Los Angeles County, Stanford Research Institute, pp. 43, 44, and 59 (1950).

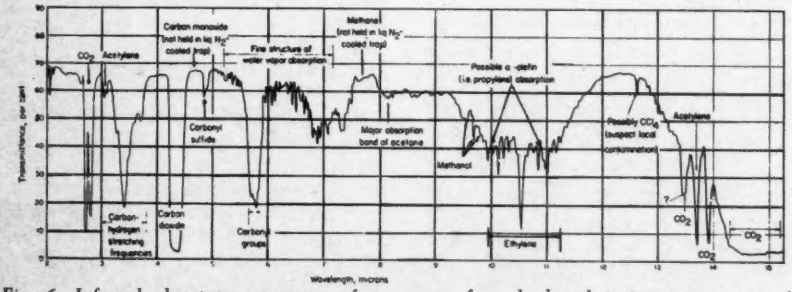

Fig. 6. Infrared absorption spectrum of raw gases from backyard incinerator run no. 17 (a composite of gases after concentration in a liquid-nitrogen-cooled trap)

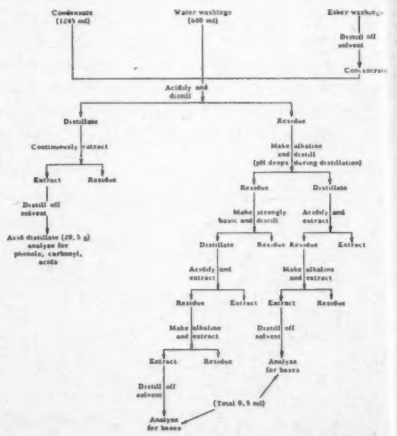

Fig. 7. A flow diagram showing the method of treating the combined condensates from runs 11-14

TABLE V

Analysis of Samples of Raw Incinerator Effluent Collected in Evacuated Flasks

	Test Run No.	Flask No.	Sampling Time After Start of Run, min.	Mg./m.³ Meter of Effluent	Lb./Ton of Charge
Ammonia (may include some low mol. wt. amines)	15	1	9-11	48	2.2
		2	30-34	30	1.4
		3	41-45	84	4.2
	16	1	5-9	28	0.9
		2	35-37	48	1.5
	17	1	7-11	28	0.9
		2	45-47	48	1.6
	Average			45	1.8
Aldehydes as HCHO	16	1	10-14	890	28
		2	39-43	595	19
	17	1	12-14	1960	64
		2	47-49	1300	43
	18	1	6-10	1660	48
		2	16-20	1110	32
		3	30-33	235	6.7
		4	45-47	182	5.2
		5	60-63	410	11.7
	Average			930	29
Nitrogen oxides at NO_2	16	1	15-19	<3	<0.1
		2	44-47	<3	<0.1
	17	1	15-17	<4	<0.1
		2	49-51	<2	<0.1
	18	1	2-5	3.2	0.09
	Average			<3	<0.1

and the relatively small number of determinations. In some cases the values are maximum figures where the amount found appeared to be below the minimum detectable limit for the compound. In other instances figures shown are minimum where it was obvious that losses of the materials in question occurred in the sampling and analytical method.

Reporting the results of absorption chemical tests for aldehydes and ammonia as averages is considered justified, since a number of determinations were made during several tests. On the basis of lb./ton of refuse burned, the average value for aldehydes (as formaldehyde) is 29 lb., and ammonia 1.8 lb. The average value for aldehydes is higher than previously reported, but that for ammonia is of the same order. Results of the analyses for nitrogen oxides is considerably lower than previously reported values.

Examination of the raw effluent gases by infrared techniques showed hydrocarbons such as methane, ethylene, benzene, olefins, and acetylene to be present in significant quantities. The same techniques showed the presence of methanol, acetone, and carbonyl sulfide. The liquid condensate contained appreciable quantities of organic acids (determined as acetic acid) and phenols.

If the weights of materials, excluding carbon monoxide, carbon dioxide, and water, are added together, a minimum-maximum range of 82 to 407 lb. of material/ton of refuse is obtained. This is a fairly wide range, but cannot be fixed more exactly without securing additional data. Earlier estimates of organic material have shown values of 145 lb. of organics from incineration of a ton of paper, and 415 lb./ton of grass clippings. All of these figures are in the same general range of values.

In addition to providing estimates of the ranges of various classes of organic compounds to be found in incinerator effluents, this work has emphasized the extreme complexity of incinerator gases. It has also pointed out the need for more work toward identifying the chemical nature of this form of pollution as a means of assessing its importance in the entire smog problem.

Acknowledgment

This study was carried on at Battelle Memorial Institute under sponsorship of the Air Pollution Foundation, Los Angeles, California. Special acknowledgment is made of the cooperation and interest of W. L. Faith, vice president, Air Pollution Foundation.

The authors gratefully acknowledge the help of J. J. Tighe and C. L. Teitelbaum, of Battelle's chemical staff, for their invaluable assistance in the analytical phases of this work.

TABLE VI

A Summary of Estimates for the Expected Ranges of Concentrations of Compounds in Incinerator Effluents

Compound or Group	Concentration, mg./m.³ of effluent	Emission, lb./ton of charge burned	Method of Analysis Used for Determination
Methanol	290-720	9-23	Infrared examination of raw effluent
Ethylene	250-1870	8-61	Ditto
Acetone	<260	>8	"
Methane	720-4600	23-150	"
Acetylene	<120-2300	<4-73	"
α-Olefins (as propylene)	<190	<6	"
Carbonyl sulfide	>80	>3	"
Benzene	>105	>3	"
Acids (as acetic)	>151	>4	Chemical analysis of liquid condensate
Phenols (as phenol)	>350	>8	Ditto
Aldehydes (as formaldehyde)	180-1960	5-64	Absorption and analysis of evacuated bottle samples
Ammonia (may include some from low mol. wt. amines)	28-84	0.9-4	Ditto
Oxides of nitrogen (as NO_2)	<3	<0.1	"
Acetaldehyde	Present	Present	Infrared analysis of materials collected in liquid N_2 cooled freeze-out trap
Esters	Present	Present	Ditto
Guaiacol	Present	Present	Infrared analysis of liquid condensate fraction
Carbon monoxide	6,250-31,400	197-990	Infrared examination of raw effluent

The Domestic Gas-Fired Incinerator's Role in Air Pollution Control

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The gas industry has been making a major contribution toward reduction of air pollution in many cities for many years, by supplying a clean-burning fuel for residential, commercial, and industrial heating and water heating. Possibly for the United States as a whole, this is the greatest single factor in the reduction of air contamination. The increasing air pollution problem of *smog* in the West, *smaze* in the Northeast, dust fall in the Midwest, industrial manufacturing effluents in the Gulf Coast Area, and all of them aggravated by a certain amount of temperature inversion appear to be the result of contributions from many other sources. The London, England Committee on Air Pollution appointed after 4,000 people were killed by the *killer fog* in December, 1952, reported[1] that "the most serious pollutants were smoke, sulfur dioxide, carbon monoxide, and grit. The domestic fire was reported to be the biggest single smoke producer." Those who have witnessed the conversion of residential heating in this country to natural gas will agree with this statement. Cleaner air and cleaner living conditions are provided by this change-over. The advantages of gas fuel in this respect were listed in my paper[2] before a section of the American Chemical Society in 1941.

Another contribution toward reduction of air pollution is being developed by the gas industry. This is the new smokeless, odorless, fly ash free, domestic gas-fired incinerator. Some day domestic gas incineration may be considered as an important companion to gas heating and air conditioning.

Opinion on the merits of present-day domestic incinerators differs greatly. Prof. G. B. Uicker,[3] after testing 14 incinerators and presenting the results of his tests recommended that the use of gas-fired domestic incinerators should not be encouraged while municipal disposal methods are studied and developed, even at the need for additional taxes. Los Angeles, California, and Allegheny County, Pennsylvania, discourage the use of domestic incinerators; in Philadelphia they are not popular; Detroit is considering very restrictive regulations; and Cleveland is granting approval for periods of 6 months at a time. The attitude of 6 air pollution officials[4] was summarized as follows:

"Domestic incinerators contribute to air pollution, although to an unknown degree. Domestic units, because of their potentially large number, present a formidable control problem. The general opinion is that satisfactory units can and will be made, and that when this is done realistic regulations can be formulated to allow their use."

To avoid any misunderstanding as to what a domestic gas-fired incinerator is designed to do, the A.G.A. definition of this appliance is: "A domestic gas-fired incinerator is sold and installed as a packaged, completely assembled, direct-fed, free-standing unit limited to not more than 5 ft.^{3} capacity and a burning rate of 25 lb./hr. of Type 1 (dry combustibles) and Type 2 (50% dry combustibles and 50% food refuse) wastes." Domestic incinerators are not designed for commercial or industrial use; they are not intended for burning large volumes of combustibles continuously; they are not backyard incinerators; they are not flue-fed, apartment house units; and they are not dependent on paper or other combustibles for incineration of food refuse.

Favorable opinion of contemporary incinerators points to unsanitary garbage cans attracting flies, rats, and vermin and to the odorous garbage trucks. At least one city, Lincolnwood, Ill.,[5] makes incinerators mandatory and several large suburbs of Cleveland require either an incinerator or a garbage grinder in all new houses. A few cities are considering making incinerators mandatory in all houses and possibly supplying the incinerators in order to reduce the expense of garbage collection and municipal incinerators or land fill. In East Ohio territory there were 111,596 incinerators installed as of January 1, 1956. In 1955 27,021 incinerators were sold. Total volume of gas used by these incinerators is estimated at 138,495,000 ft.^{3}/mo. with an aggregate revenue for the gas company of $1,047,011.40/yr. and cost to the customer of 78.75¢/mo. Incinerator sales amounted to about $11,159,600.00 in the past 5 yr. Certain large housing developments in our area have incinerators in every house. No odor or smoke complaints are received from them. Surveys have shown that owners are well-pleased with the incinerators.[6,7] Such complaints as have been

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[1] Smog Scramble Spans Nation. *Chem. & Eng. News*, 32, 1113 (March 22, 1954).

[2] F. E. Vandaveer and W. L. Jones. City Gas—The Ideal Smokeless Fuel for Domestic and Industrial Uses. *J. Ind. & Eng. Chem.*, p. 852 (July 1941).

[3] G. B. Uicker. Survey of Gas-Fired Domestic Incinerators. *J. Air Poll. Control Assoc.* 58, 199 (Feb. 1956).

[4] J. S. Boylan and H. W. Nelson. Survey of Odor and High Temperature Problems of Domestic Gas-Fired Incinerators. Battelle Memorial Institute. Rept. to A.G.A. Committee on Incinerator Research. (Dec. 10, 1953).

[5] In Lincolnwood, Illinois, Incinerators Make the Difference. *Gas Appliance Merchandising*. p. 10 (July 1955).

[6] F. E. Vandaveer. Gas Incinerator Characteristics, Acceptance, Servicing and Load. *A.G.A. Monthly* p. 33 (Oct. 1954).

[7] F. E. Vandaveer. Domestic Gas-Fired Incinerators. *J. Air Poll. Control Assoc.* 5, 197 (Feb. 1956).

received from neighbors can usually be corrected by educating the owner to wrap the garbage with more paper and to shake the grates and empty the ash receptacle more often.

A. G. A. Research on Incinerators Started

Despite the good reception and good performance of contemporary incinerators, it was recognized in 1952 that improvements could be made and that an opportunity existed to make incinerators smokeless, odorless, and fly ash-free. A research program designed to accomplish this purpose was started by the American Gas Association in July, 1953. About the same time the A.G.A. Approval Requirements Committee on Incinerators began extensive revision and strengthening of the requirements on domestic gas-fired incinerators. Certain incinerator manufacturers also increased their research and development program. Within a time span of 2½ yr. great progress, as briefly outlined below, has been made.

Development of Test Charges and Test Methods for Odor, Smoke, and Fly Ash

For test and research purposes it was necessary to develop a smoke-and-odor-producing charge and a high temperature and fly ash-producing charge. Many types of mixtures, including household and restaurant garbage, various foods, cake, bread and an assortment of papers and cardboard were tried. The most odorous charge seemed to be wet garbage. Hair, vacuum cleaner sweepings, and feathers also produce objectionable odors, but these items are very infrequently added to the domestic garbage load. Finally, a mixture of 60% food refuse containing potatoes, cabbage, oranges, bread, rice, beef suet, water, and 40% combustibles consisting of corrugated cardboard, newspapers, and waxed paper was selected. The exact composition is given in A.G.A. Approval Requirements for Domestic Incinerators, effective January 1, 1956. For a high temperature, fast burning, fly ash-producing charge, shredded paper was selected after comparative tests with excelsior, wood blocks, crumbled newspaper, cardboard, and heavy wrapping paper. Seven or more charges of 2 lb. of shredded paper at 8 min. intervals is far more severe as a test charge than would be encountered in service. The garbage mixture produces plenty of odor and fly ash and it simulates the waste material from residences very well.

Test methods and instruments for determining smoke, odor, and fly ash in the flue gases from incinerators have also been developed. The incinerator is arranged for test as shown in Fig. 1. For determining odor 3 observers compare the odor from burning 2 sheets of newspaper with the full discharge of flue gases from the incinerator for a few seconds into the room while burning the garbage mixture. An odor stronger than the smell of burning newspaper is not tolerated. This method was selected as practical, reasonably accurate, and easily duplicated. Various odor testing procedures given in the literature were studied and the most likely methods were tried, including a constant flow sampling and air dilution method, an odorimeter, an osmometer, and chemical analyses for aldehydes. This subject of odor determination alone could have developed into a lifetime project.

For determination of smoke and fly ash and to some extent odor, a Bacharach True Spot Smoke Tester was selected. This instrument is used in testing for smoke from oil burners. It is equally applicable to incinerator smoke. Even a thin white smoke will stain black, the filter paper through which the flue gases

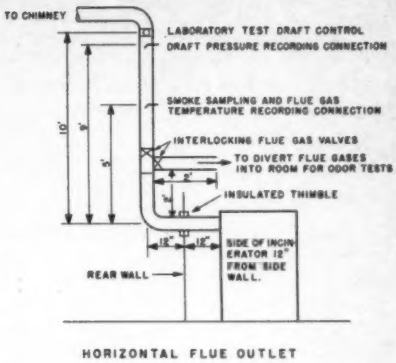

TO CHIMNEY
LABORATORY TEST DRAFT CONTROL
DRAFT PRESSURE RECORDING CONNECTION
SMOKE SAMPLING AND FLUE GAS TEMPERATURE RECORDING CONNECTION
INTERLOCKING FLUE GAS VALVES
TO DIVERT FLUE GASES INTO ROOM FOR ODOR TESTS
INSULATED THIMBLE
SIDE OF INCINERATOR 12" FROM SIDE WALL.
REAR WALL
HORIZONTAL FLUE OUTLET

Fig. 1. Arrangement of apparatus for incinerating effectiveness, smoke, odor, and fly ash tests and flue gas temperature tests.

are pulled. Both the automatic and the manual samplers are shown in Fig. 2. The proposed requirement is to allow not more than Bacharach No. 4 smoke scale for 10 min. in 24 hr. during the burning of the garbage mixture and the shredded paper. Additional tests for fly ash are to be made with a Bausch and Lomb dust counter as shown in Fig. 2, or by collection on a sticky plate and subsequent measurement of size and number of particles with a microscope and measuring scale.

Fundamentals of Complete Incineration

During experimental work on this research project certain fundamental principles were kept in mind. *i.e.*, when garbage and paper burn completely to carbon dioxide, water vapor, and ashes there is no odor in the flue gases. Odor is generated by partial, incomplete combustion and by destructive distillation

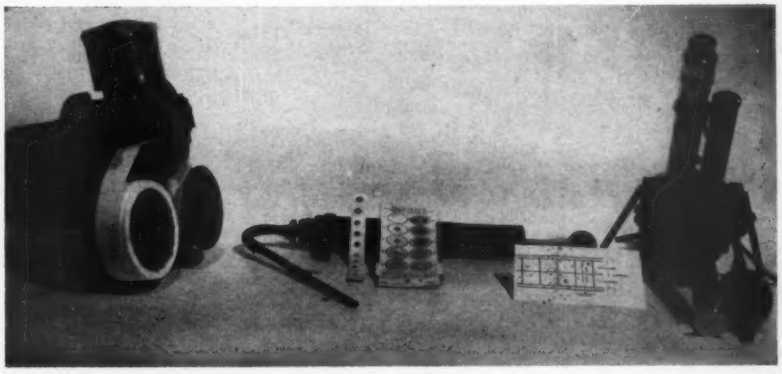


Fig. 2. Instruments selected for determining smoke and fly ash. At left, Bacharsch automatic continuous true spot smoke tester, showing roll of filter paper through which flue gases are pulled for 25 sec. (112 in.² of sample) for each spot; it will record one or two spots/min. as desired; pump not shown. In center, Bacharach true spot smoke tester (manual pump), the smoke scale (0-9) and typical spots from white smoke. At right, Bausch and Lomb dust counter, and enlarged scale and dust spots as seen through the microscope of the instrument.

and cracking of the combustibles. Garbage containing sulfur and nitrogen compounds will form sulfur dioxide and nitric oxide in extremely small amounts and these gases have an identifiable odor. Probably the most objectionable odors produced by incomplete burning of garbage are the aldehydes, dialdehydes, and ketones with sharp, unpleasant, rancid smells, and the organic hydrocarbons and organic acids with sharp irritating smells. Production of odor or smoke from incineration of garbage may be attributed to 1 or more of the following conditions: (1) Reaction temperature below the ignition temperatures of the vapors; (2) Insufficient oxygen supply for combustion at the right place; (3) Removal of these vapors from the reaction zone before completion of the combustion reaction. Complete incineration must be done in 2 stages, although both stages may proceed simultaneously or in very rapid succession once ignition occurs. In the first stage, drying, ignition, and combustion takes place, progressively converting part of the solid charge into a gaseous or vapor state, burning of most of the gases and leaving an ash residue on the grates. In the second stage total oxidation of the volatile products, odor, and smoke escaping from the first stage must be accomplished. All present day domestic incinerators have the first stage of incineration and varying amounts of the second stage, but none of them have a complete second stage of burning. The major task of this research project, accordingly, was to develop practical, economical methods of consuming smoke and odor and producing no fly ash.

Second Stage Incineration Accomplished by Several Methods

Several satisfactory methods to accomplish second stage incineration were found and tests were made to verify them.

Catalysts, platinum or palladium, impregnated on porcelain or chrome alloy heated to 500-700°F and supplied with air for the combustion reaction will consume all smoke and odor. The combustion reaction temperature is lowered as much as 600°F with catalysts. To prevent escape of smoke or odor through catalysts they must be heated to the proper temperature and maintained at that temperature by a gas flame, and the construction must be such as to cause intimate contact of flue gases with them. Fly ash deposits and poisonous vapors of metals must be kept from the catalysts, or provision must be made to purge and clean the catalyst each time with a gas flame before the first stage incineration starts. This can be done. Adequate air supply for combustion on the catalyst, control of rate of incineration, and maintenance of catalyst temperature are other requisites of design.

Gas heated ceramic balls,[8] or slabs[9] raised to a temperature of 1400-1500°F will burn all smoke and odor if adequate air is available at that location. It is not sufficient to have a temperature just above the ignition temperature of approximately 1100°F for most organic vapors.

Gas-heated stainless steel grids heated above 1200°F will consume all smoke and odor. Proper design of the grid, selection of proper composition of steel to withstand the temperatures involved, and good application of gas flame to the grid will provide smokeless incineration.

The smokeless combustion principle as applied to a coal stove as developed by Battelle Memorial Institute several years ago was also tried on incineration of garbage. This work was done at Battelle and was sponsored by the American Gas Association. This principle of downdraft combustion was successful when it was augmented by a gas burner to both speed up the incineration and an after burner gas flame to consume the smoke and odor which escapes from the first stage.

Flame of a gas-fired after burner can destroy all smoke, odor and fly ash from burning garbage and combustibles. It has been known for a long time that smoke passing through a flame is destroyed. This is demonstrated in smokeless gas range broilers, and in smoke passing through the flame of a match or cigarette lighter. The trick in incineration of garbage is to keep the gas flame burning in an atmosphere of flue gases, smoke, and ashes. Air has to be introduced at the right place, the burner has to be properly located, burning rate of combustibles has to be controlled, updraft has to be limited to prevent fly ash entrainment, adequate safety controls must be used, flue gas temperature must be held below 1400°F, and all this done within a space about the same size as contemporary incinerators, and at a price that will be attractive.

[8] L. T. Bissey. New Design Household Incinerator Eliminates Smoke and Odor. *Gas Age*, p. 30 (1948).

[9] American Gas Association Research Project DA-1-M on Elimination of Smoke, Odor and Potential High Temperature Hazard From Domestic Gas-Fired Incinerators.

A.G.A. Laboratories and Battelle Memorial Institute Prototype Models of Smokeless, Odorless, Fly Ash Free Incinerators

Two prototype smokeless, odorless, and fly ash-free incinerators developed by A.G.A. research will now be described. These models have not been subjected to detailed testing or field trials as yet. Predecessor models have complied with most of the proposed new A.G.A. requirements for incinerators. These models are described to illustrate the principles involved and to show that satisfactory incineration can be accomplished.

A model as developed by Battelle Memorial Institute[10] is shown in Fig. 3. This incinerator is the result of many tests, many changes, consultations, and good experimental work. It has downdraft combustion, a combination gas-fired incineration burner and after burner, restricted supply of preheated air, 6 in. flue collar with adjacent barometric draft control, grate sloping toward burner, a safety vent when door is open, and stands about 47 in. high, 22 in. deep, and 22 in. wide. Capacity is 1.5 bu. and gas input rating is approximately 30,000 Btu./hr.

[10] Research Done by G. M. Hein with Dr. Harlan W. Nelson, Battelle Memorial Institute, Columbus, Ohio.

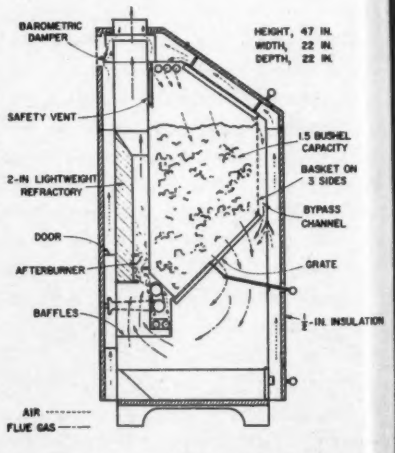


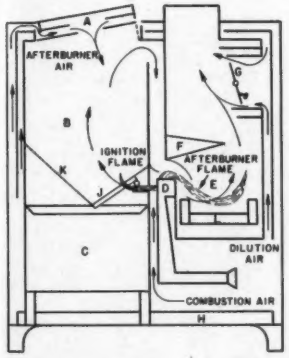
Fig. 3.

The A.G.A. Laboratories prototype model[11] is shown in Fig. 4. It is the result of a great deal of experimental work and application of good engineering principles. It employs updraft combustion of garbage, downdraft combustion of smoke and odor in a gas flame after burner with ceramic baffles in the hot second stage incineration chamber. Restricted air for combustion, preheated downdraft air for after burning, grate sloping toward burner, large ash receptacle, a large settling chamber for fly ash and a barometric damper are other features. The unit has 2 automatic pilots, a timer, a pressure regulator, and a gas cock. Capacity is 1.5 bu., input rating 35,000 Btu./hr. with about 10,000 Btu. to the first stage burner and 25,000 Btu. to the after burner. Dimensions are approximately 40 in. high, 31 in. deep, and 18 in. wide.

Supplementary Fuel, Gas, Required

A supplementary fuel, such as gas, is required to incinerate garbage in a 1-stage incinerator within a reasonable time. Tests have shown that the amount of gas fuel needed is from 2,050 to 8,640 Btu./lb. of garbage. To eliminate smoke and odor from flue gases, tests have indicated a requirement of 2½ to 3 times as much gas heat for the second stage incineration as for the first stage. A gas burner in this second stage is essential and the input rating of such a burner for a 1.5 bu. incinerator would be from 18,000 to 30,000 Btu./hr.

[11] Research done by D. W. Skipworth and A. L. Nead with Walter B. Kirk, A.G.A. Laboratories, Cleveland, Ohio.



A- CHARGING DOOR
B- INCINERATION CHAMBER- 1ST STAGE INCINERATION.
C- ASH RECEPTACLE
D- BURNER
E- AFTERBURNER CHAMBER- 2ND STAGE INCINERATION
F- FLUE GAS DEFLECTOR
G- DAMPER
H- BASE
J- HINGED GRATE
K- HOPPER
L- CERAMIC BRICK

Fig. 4. A.G.A. Laboratories Prototype Incinerator for A.G.A. DA-1-M

TABLE I

Data on Four Research Models of Incinerators for Smoke, Odor, and Fly Ash Elimination During Incinerating Effectiveness Test (Garbage and Combustibles)

Research Model	1st Day	2nd Day	3rd Day	4th Day	Result of Test
Smoke Tests (Not Over Bacharach Smoke Scale No. 4 for 10 Min.)					
1	O	O	O	O	OK
2[a]	#5-8 for 14 min.	#4-8 for 12 min.	#4-8 for 21 min.	O	Fail
3	#4 for 1 min.	#4-9 for 6 min.	#5 for 1 min.	#4-6 for 11 min.	Almost OK
4	#4 for 2 min.	O	O	#4 for 2 min.	OK
Odor Tests (3 Observers Comparing With Odor From 2 Newspaper Sheets Burning)					
1	Less or like	Less or like	Less or like	Less or like	OK
2	More	Much more	More	More	Fail
3	OK but slight smoldering	OK but smoldered for one hour	OK but smoldered for one hour	OK but smoldered for one hour	OK if smoldering is corrected
4	OK	Less to more	Less to slightly more	Less to more	OK—but on line
Fly Ash (Not Over 150 μ)					
1	OK (40 μ)	OK (60 μ)	OK —	OK (75 μ)	OK
2	OK (50 μ)	OK (45 μ)	OK (90 μ)	OK —	OK
3	OK (60 μ)	OK (45 μ)	OK (45 μ)	OK (65 μ)	OK
4	OK (65 μ)	OK (70 μ)	OK (90 μ)	OK (150 μ)	OK
Smoke Discharge at Door (Visible During Test)					
1	No	No	No	No	OK
2	Yes (6 min.)	O	O	Yes (7 min.)	Fail
3	Yes (1 min.)	Yes (4 min.)	Yes (2 min.)	O	Easily corrected
4	No	No	No	No	OK

[a] This unit was not ready for this test. It has since been greatly improved.

A.G.A. Approval Requirements Greatly Strengthened

Improvements in A.G.A. Approval Requirements for Domestic Gas-Fired Incinerators have kept pace with research and development[12]. Ten major changes appeared in the January, 1956, requirements which were not in the 1953 requirements. These included a reduction in updraft during the test from .03 in. to .01 in., required operating and installation instructions, pilot filters, flue gas temperatures limited to less than 1400°F, ash receptacle size required was doubled, approval on natural gas only as an optional feature, a new garbage-paper charge simulating household refuse and charging over a period of 4 days' time similar to domestic usage, and smoke limited to not exceed 1½ Ringleman for 8 min./day. Ten more major changes are proposed, pending field tests on new incinerators this year, for effective date of January, 1958. In those requirements it is proposed to require the 4-day garbage charge to be so completely burned as to leave only 5 oz./bu. of incinerator capacity of uncarbonized material, smoke and fly ash shall not exceed Bachrach smoke scale No. 4 for 10 consecutive min., odor at any time shall not exceed that of burning 2 newspaper sheets as determined by 3 observers, fly ash particles shall not exceed 150 μ, and no observable smoke shall escape into the test room from the incinerator doors or joints. No contemporary incinerator will meet these requirements. However, 4 research models subjected to these proposed tests yielded results as shown in Tables 1 and 2. Two of the incinerators were A.G.A. research models and 2 were models made by 2 manufacturers. Units 1 and 4 passed all tests, No. 3 would comply if the smoldering and smoke discharge at the door was corrected, and Unit No. 2 failed on several tests as it was obviously not ready for these tests—it has since been redesigned, and pre-

[12] F. E. Vandaveer. Domestic Gas-Fired Incinerators, Present and Future, A.G.A. Research and Utilization Conference, Cleveland, O. (Apr. 3-4 1956).

TABLE II

Data on Four Research Models For Smoke, Odor, and Fly Ash Elimination During Fire Hazard Test (Shredded Paper Burning)

Unit	Bacharach Smoke Scale No.	Duration—Minutes	Result of Test
Smoke Tests (Not Over No. 4 Bacharach Smoke Scale for 10 Min.)			
1	4 to 7	3	OK
2[a]	4 to 9	Most of time	Fail
3	4 to 8	2	OK
4	4 to 6	6	OK
Odor Tests (Three Observers Comparing With Odor From Two Newspaper Sheets Burning)			
1	Like		OK
2	Like		OK
3	Like		OK
4	Like		OK
Fly Ash (Not Over 150 μ)			
1	Clean		OK
2	OK (45 μ)		OK
3	Clean		OK
4	75 μ + 1 large particle		OK
Smoke Discharge at Door (Visible During Test)			
1	Slight		OK
2	O		OK
3	O		OK
4	O		OK

[a] This unit was not ready for this test. It has since been greatly improved.

liminary tests indicate probable compliance with these proposed requirements.

Manufacturers' Field Trial Models of Smokeless, Odorless, Fly Ash Free Incinerators*

Two manufacturers, have reached the stage in their development work where each company intends to make and sell about 1,000 smokeless, odorless, and fly ash free incinerators in 1956. If these field tests are successful greater production will be undertaken. At least 4 other companies have advised me that they are well-advanced with their new models and will attempt to comply with the proposed new A.G.A. requirements. There is no doubt that manufacturers' research and development was stimulated by American Gas Association research, by the interest and increasing sales created by certain gas companies, by the large potential market for new units, by the pressure of restrictive air pollution regulations, and by city officials.

* Author will identify units upon request.

Incinerator (A)

A drawing of new incinerator (A) is shown in Fig. 5. The round tube across the bottom of the grate to the back has the burner inside of it and is open on the bottom side. The radiation from this tube causes the refuse to ignite and burn and the smoke and flue gas travel is downward through the combustion area and into the bottom of this horizontal round pipe which is open on the bottom. Since the pipe is completely filled with fire and the rectangular box at the back for the 90° turn is also full of fire, the smoke and odor are burned before they rise through the stainless steel pipe in the back and reach the outlet of the incinerator. The square box on the back of the incinerator houses a barometric type control which not only controls the air inlet to the top of the combustion area but also allows secondary air to enter into the smoke stack in order to keep it cooled as the heat and draft increase. The controls are to be mounted in a housing similar to that shown on the front. The early models will have a timer than can be manually set to different lengths of time but with instructions to set it to the full period. A control is being developed so that it has to be turned on all the way and will make the complete time cycle under all conditions.

Incinerator (B)

New incinerator (B) smokeless, odorless, and fly ash-free, is shown in Fig. 6. The burning cycle is controlled by a Duomatic timer valve with *on* and *off* positions. For 20 min. the timer allows gas to flow and burn at the after burner thereby preheating and purifying the catalyst bed, and establishing a good draft to accept the burning cycle and increase the air flow through the combustion chamber. After the preheating time elapses, a cam actuates a valve slide allowing gas to flow to the first stage incineration burner (8,000 Btu./hr.) and at the same time decreases the flow to the after burner from 18,000 Btu./hr. to approximately 16,000 Btu./hr. Corrugations in the combustion chamber walls allow air to circulate around the refuse, prevents refuse build-up along the sides, and provides circulation of air and flames along the sides thereby aiding in destruction of the refuse. The flue gases pass upward through the rear grill, through the flames of the second stage burner and through the heated catalyst (at about 1000°F) where all unburned gases are scrubbed against coated saddles and burned to CO_2 and water vapor. The flue gases pass into the flue pipe where they are diluted with air through the barometric draft control. The draft control is set to provide an updraft of .035 in. to .04 in. water column and to drop the flue gas temperature to about

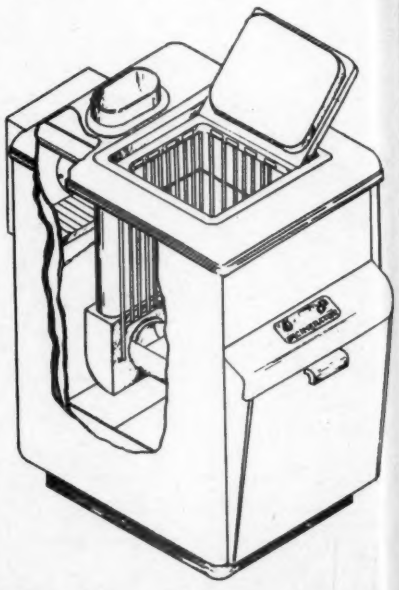


Fig. 5. New smokeless-odorless incinerator.

750°F. Air for combustion and incineration is supplied under each burner and for second stage combustion by downdraft air flowing up between jacket and combustion chamber walls and down through the liner of the door. Incinerator (B) is equipped with a timer, 2 automatic pilots, 2 burners, a pressure regulator, a barometric draft control, and large ash receptacle. Gas consumption is estimated at 70 ft.³/day for 1 complete cycle of 1 hr. and 20 min., including 2 pilots at 800 Btu./hr. each. On 63¢/1000 ft.³ gas, the operating cost would be $1.32/mo. Incinerator (B) is 36 in. high, 24 in. deep, and 18 in. wide. It will be in limited production within the next few weeks.

Trace Constituents (Nitrogen Oxides, Sulfur Dioxide, Aldehydes) Below Acceptable Standards

In any discussion of air pollution contribution from combustion or incineration equipment the question of trace constituents in the flue gases will arise. Although it has been shown that with reasonable care in operation the odor, smoke, and fly ash from these new incinerators will be acceptable and practically non-existent, the question may arise about nitric oxide, sulfur dioxide, and aldehydes or organic material. There has not been time enough to date to make specific tests and analyses for these trace constituents during the A.G.A. research on the new incinerators. However, calculations using published data on municipal incinerators and gas appliances will be given. These calculated values are probably as good as could be obtained by actual test. Data supplied by one of the manufacturers will also be presented.

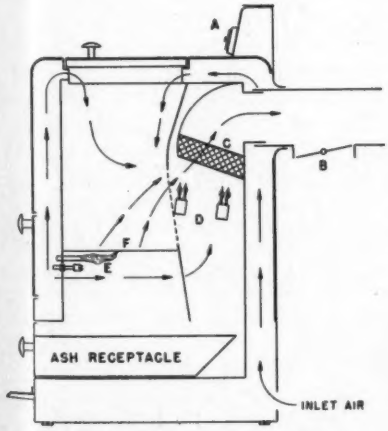


Fig. 6. New smokeless-odorless incinerator.

TABLE III

Maximum Allowable Concentration of Possible Flue Gas Constituents From Incinerators (8 Hr. Exposure, No Effect on Health) and Concentration of These Constituents in 5 Cities

(Maximum Allowable Value (except water vapor) Adopted by Conference of Governmental Industrial Hygienists—1953)

Constituent	Maximum Allowable ppm.	Concentration of air of 5 cities PPM	
		Range	Mean
Water vapor (saturated air)	17,400+	—	—
Carbon dioxide	5,000	—	—
Carbon monoxide	100	—	—
Nitrogen dioxide	10	0.0 — 3.48	.10 — .89
Nitrogen oxides (other)	25	—	—
Formaldehyde	5	0.0 — 1.0	.067 — .20
Sulfur dioxide	10	0.0 — 1.45	.023 — .20

The maximum allowable concentration of gases which might be present in flue gases from incinerators which can be breathed continuously for 8 hr. without injury to health and the concentration of these constituents in the atmosphere of five cities has been listed by Katz[13] and is given in Table III.

It has been reported that nitric oxide in the flue gases from burning gas in domestic appliances may vary from 0 to 50 ppm.[14] One of these new incinerators burning 70 ft.³ of gas/day would possibly produce .0735 ft.³ of NO_2. Assuming there is an incinerator in every house in a city of 500,000 houses, that the area involved is 20 miles long by 12 miles wide (similar to Cleveland, Ohio), and an air height of 50 ft. for calculating purposes, there would be 335 billion ft.³ of air in such an area and the nitrogen dioxide concentration would never exceed 0.1 ppm. If an air height of 1,000 ft. is taken, the concentration would never exceed .005 ppm. This is assuming uniform distribution of the gas in this air volume and no air movement out of or into this area.

Aldehydes have been reported[14] as possibly varying from 0-11 ppm. in flue gas from gas appliances. Under the same conditions listed above for nitrogen dioxide the concentration in the air above a city of 500,000 houses would never exceed .02 to .001 ppm.

Sulfur dioxide, assuming 0.1 gr. of sulphur/100 ft.³ of natural gas from the odorant used, would contribute a maximum of .016 to .003 ppm. to the atmosphere of this same city under the same conditions.

From the incineration of garbage and paper what would be the addition of nitrogen dioxide, sulfur dioxide, and aldehydes to the atmosphere? In a booklet on municipal incineration[15] it is stated that daily production of garbage/person is about ⅔ lb., and of combustible refuse about 2 lb./person. Assuming a family of 4 the amount of garbage and combustibles/day would be about 11 lb. Also, in municipal incineration[15] it is stated that stack gases from a good municipal incinerator burning municipal refuse contain about 1.1 lb. of aldehydes/ton of refuse, 2.4 lb. of nitrogen oxides, and 1.9 lb. of sulfur dioxide. Relating these values to a single domestic incinerator, assuming the new domestic incinerator to be as good or better than the municipal incinerator, burning 11 lb. of refuse/day a total of approximately .112 ft.³ of NO_2, .063 ft.³ of SO_2, and approximately .0505 ft.³ of aldehydes would be liberated. Again using the example of an incinerator in each of 500,000 houses the air over that area to a 50 ft. height, assuming no air movement whatever except mixing of the gases within the area, would contain 0.16 ppm. of NO_2, .009 ppm. of SO_2, and .074 ppm. of aldehydes. Adding together the trace gases generated from both gaseous combustion and refuse incineration, the total would be .26 ppm. of NO_2 at the 50 ft. height, .025 ppm. of SO_2, and .094 ppm. of aldehydes. If mixing occurred to a 1000 ft. height the numbers would be .013 ppm. of NO_2, .001 ppm. of SO_2, and .005 ppm. of aldehydes.

Comparing the above data with the maximum allowable amounts and the present concentration in air over 5 cities

[13] Morris Katz. Recent Developments in Atmospheric Pollution, *A.G.A. Proc.* p. 796 (1954).

[14] Edwin L. Hall. What is the Role of the Gas Industry in Air Pollution. *Gas*, p. 54 (Oct. 1952).

[15] Municipal Incineration, Sanitary Eng. Research Project. *Univ. of California Tech. Bull.* No. 5, pp. 17, 71, and 56 (June 1951).

as given in Table III, it is readily apparent that incinerator additions, assuming the impossible of an incinerator in every house of a 500,000 house city, would be insignificant and far below the allowable values. In most locations in this country there is some air movement at all times or for most of the day. This air would dilute the amounts listed above to fantastically low figures. Still further dilution would occur since the ceiling height of air over a city is far greater than the maximum figure of 1,000 ft. used in these calculations.

As a direct indication of what one new domestic incinerator will do in burning up smoke, odor, and particulate matter results of tests compared with published data on municipal incinerators are given in Table IV.

The tests were made during the burning of 6 lb. of garbage-paper mixture per day (A.G.A. test charge plus domestic garbage); gas consumption was 29.4 ft.^{3}/charge, flue gas volume was 4,817 ft.^{3}, flue gas analysis top of stack was 8% CO_2, 13% O_2, 0.0% CO. Analyses were made by the best-recognized methods available. Attempts are being made by the manufacturer to reduce these trace constituents still further. It will be apparent, however, that the new domestic incinerator is considerably better than the reported results on municipal incinerators, which are generally regarded as satisfactory by city officials.

There should be no necessity for presenting data herein comparing the flue gases from these new domestic incinerators with flue gases and exhaust gases from other solid or liquid burning appliances. Air pollution officials are familiar with published data on automobile and truck exhaust gases, smoke and flue gases from coal and oil appliances, from industrial boilers burning coal, from electric generating stations burning coal, and from municipal incinerators. These new domestic incinerators, if operated properly, the grates shaked regularly and ashes removed when receptacle is nearly full should contribute far less to air pollution than any other combustion source, except possibly domestic gas appliances.

Incinerator Ash Analysis

Analysis of the ash remaining from domestic incinerator tests has not been reported to date, but, undoubtedly, it would be similar to that from municipal incinerators. There are practically no unburned materials left on the grates of these new incinerators after the incineration process. The following analysis of ash was obtained from municipal incineration.[15]

It will be apparent from Table V that there is no nitrogen or oxides of nitrogen in the ash but the items listed, particularly phosphates and potash, are good for certain fertilizer purposes. However,. it is not a complete fertilizer. It is probably best to dispose of the ashes by dumping or putting them with the noncombustible refuse.

Summary

(1) The gas industry has been making a major contribution toward air pollution control for many years by supplying a clean burning fuel for heating purposes.

(2) Another contribution toward air pollution abatement, the new smokeless, odorless and fly ash free domestic gas-fired incinerator, is being developed by the gas industry.

(3) A.G.A. research on incinerators was started in July, 1953 with the objective of developing methods for eliminating smoke, odor, fly ash, and high temperature hazards from domestic gas-fired incinerators.

(4) An odor and smoke producing charge of garbage and paper, and a high temperature and fly ash producing charge (shredded paper), have been developed for test purposes.

(5) Test methods for determining odor (three observers comparing flue gas with odor of burning newspaper), smoke (Bacharach true spot smoke tester), and fly ash (Bausch and Lomb dust counter) have been selected.

(6) Two-stage incineration with gas heat in both stages is necessary to eliminate smoke and odor from domestic incinerators; the first stage burns the garbage and paper; the second stage oxidizes the smoke and odor escaping from the first stage. The second stage burner requires from 2½ to 3 times (18,000-30,000 Btu./hr.) as much heat as the first stage burner for a 1.5 bu. incinerator.

(7) For second stage incineration, catalysts impregnated on porcelain or on chrome alloy heated to 500-700°F, ceramic balls or slabs heated to 1000-1500°F., stainless steel grids heated above 1400°F, smokeless downdraft combustion with a gas after burner, and the flame of a gas-fired after burner may be used.

(8) Drawings of 2 prototype models of smokeless, odorless and fly ash free incinerators developed by American Gas Association research sponsored at A.G.A. Laboratories and Battelle Memorial Institute and test data on them are presented.

(9) Ten major changes and additions to A.G.A. Approval Requirements for Domestic Incinerators have been made for 1956 and 10 more are proposed for 1958 which, if adopted, will insure smokeless, odorless, and fly ash-free incinerators. No contemporary incinerators could comply with these new requirements.

(10) Two manufacturers intend to market approximately 1,000 new incinerators each this year for field trial. One

[15] See footnote 15, page 95.

[16] Private Communication from Calcinator Corporation, March 10, 1956. Data obtained from commercial testing laboratories.

[17] Municipal Incineration. *Univ. of California Tech. Bull.* No. 5, p. 71 (June 1951).

[18] Municipal Incineration. *Univ. of California Tech. Bull.* No. 6, p. 31 (Nov. 1951).

TABLE IV

Stack Discharge Anaylses for Trace Constituents in a New Domestic Incinerator Compared With Municipal Incineration

	New Imperial[16] ppm.	Municipal Incinerator[17] ppm.
Oxides of nitrogen	1.60	110
Organic acids	2.2	51
Aldehydes (as formaldehyde)	19	60
Particulate matter	.48 gr./ft.^{3}	.95 to 2.31 gr./ft.^{3} without[18] spray chamber; .164 to .564 gr./ft.^{3} with spray chamber.

TABLE V

Chemical Analysis of Ash Cincinnati, O. Incinerator, 1947

Sampling Point	Phosphoric Acid (P_2O_5)	Potassium Oxide (K_2O)	Calcium Oxide (C_aO)	Magnesium Oxide (M_gO)	Manganese	Copper	Unidentified Ash
Ash	1.32%	1.9%	14.80%	1.74%	.004%	.08%	79.4%
Fly Ash	1.51%	4.47%	12.30%	.48%	.002%	.006%	81.23%

of these models has a gas-heated catalyst in the second stage; the other employs downdraft combustion and a combined incineration burner and gas flame after burner. Drawings of both models are presented herein.

(11) Calculations have been made from published data showing that an incinerator in every house in a large city should not cause an increase in nitrogen oxides, sulfur dioxide, or aldehydes in the atmosphere to anywhere near the allowable maximum level for health.

(12) Test data presented by 1 manufacturer company shows that nitrogen oxides, aldehydes, organic acids, and particulate matter from its new incinerator is considerably lower than is normally obtained from municipal incinerators. Municipal incinerators, properly designed and operated, are generally regarded as contributing very little to air pollution. There is reason to believe that the other 3 research models will do as well as the one on which tests have been made.

(13) Ashes from municipal incineration contain phosphates, potash, calcium and magnesium oxides, but no nitrates. Although they would be good for some soils, they are not complete fertilizers. Similar ash would be expected from these new domestic incinerators.

(14) If the field test program on new incinerators during 1956 is successful, plans have been proposed to make the proposed new A.G.A. requirements effective in 1958, and possibly to require all incinerators approved and produced after January 1, 1959 to comply with these new requirements. This latter date and condition are subject to action by other A.G.A. committees and Gas Appliance Manufacturers' Association approval.

(15) To progress from a research idea to prototype models, to manufacturer field trial models, and to complete new A.G.A. requirements on incinerators in $2\frac{1}{2}$ years is a remarkable achievement. It was accomplished by the cooperation of many people and a number of companies and institutions.

Thermodynamic Considerations in the Interactions of Nitrogen Oxides and Oxy-Acids in the Atmosphere*

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It is well known that oxides of nitrogen exist in urban atmospheres. Particularly high concentrations of nitrogen oxides have been reported over the coastal cities of California [1]. These gases are formed in part by high temperature reactions between nitrogen and oxygen to form nitric oxide, NO, in the combustion processes occuring in automobile, truck and bus engines and in furnaces, heaters and incinerators.

Actually, a large number of solid, liquid and gaseous nitrogen oxides are known to chemists. Only those known or postulated as existing in the gaseous state will be considered in this discussion.

The nitrogen oxides which have been reported to exist in the gas state include N_2O, nitrous oxide; NO, nitric oxide; NO_2, nitrogen dioxide; NO_3, nitrogen trioxide; N_2O_3, dinitrogen trioxide; N_2O_4, dinitrogen tetroxide; and N_2O_5, dinitrogen pentoxide. (Table I). An absorption spectrum presumed to be nitrogen trioxide, NO_3, has been observed when nitrogen dioxide, NO_2, is reacted with ozone, O_3. The concentration of NO_3, if it exists, is so low that it can be ignored as an atmospheric contaminant. Nothing is known about the concentration of N_2O_5 in the atmosphere. If the mechanisms which have been advanced to explain smog in California are at all correct, N_2O_5 must have some unknown but quite small steady state concentration. It will be shown later that of the total concentrations of nitrogen oxides existing in the atmosphere, the concentrations of N_2O_3 and N_2O_4 may also be expected to be very small.

Thus, the nitrogen oxides likely to be present in any substantial quantity in the atmosphere are only nitrous oxide, N_2O, nitric oxide, NO, and nitrogen dioxide, NO_2. Nitrous oxide is stable and neither decomposes nor oxidizes except at high temperatures. It becomes toxic only in very high concentrations. However, its existence in the atmosphere and the way in which it is produced are of considerable interest. Both nitric oxide and nitrogen dioxide exist in pphm. to ppm. concentrations in urban atmospheres and are believed to play important roles in the formation of smog. In addition to the nitrogen oxides, there are 2 oxy-acids, nitric acid and nitrous acid, which can exist in urban atmospheres in the form of aerosols.

TABLE I

Estimated Concentrations of Gaseous Nitrogen Oxides and Oxy-acids

Nitrogen Oxide or Oxy-acid	Concentration in Urban Atmospheres
N_2O	ppm. range (0.5 ppm.)
NO	pphm. to ppm. range
NO_2	pphm. to ppm. range
NO_3	very small
N_2O_3	very small $< 10^{-6}$ ppm.
N_2O_4	very small $< 10^{-6}$ ppm.
N_2O_5	small
HNO_2	small
HNO_3	ppm. range(a)

(a) Thermodynamic and kinetic considerations indicate ppm. of HNO_3 could exist, but experimental values are not available.

Equilibrium constants allow the calculation of the equilibrium concentrations of various materials entering into reaction with each other. The equilibrium constants reported herein have been specifically calculated from recent spectroscopic and structural measurements.

It must be realized that the equilibrium conditions to which the data apply may be reached very slowly or very rapidly, or at some intermediate rate. Determination of this rate is a problem for experimental chemical kinetics rather than thermodynamics.

Thermodynamic information can tell whether a small or large amount of a given reactant will exist at equilibrium. For example, if the equilibrium constant is such that only very small concentra-

[1] Clean Air for California. Initial Report of the Air Pollution Study Project, State of California, Dept. of Public Health.

* Presented at the 49th Annual Meeting of APCA held at Buffalo, N. Y., May 20-24, 1956.

tion of the products of reaction are present at equilibrium at the temperatures of interest, then it does not matter whether the reaction proceeds slowly or rapidly because there will not be much of the material produced by the reaction regardless of reaction rate. On the other hand, if the equilibria favors the products of reaction, it is very important to know the rate at which the reaction proceeds.

Finally, in some reactions the presence of light of certain wave lengths is very important. Such photochemical reactions depend on the efficiency of utilization of light energy at a given wave length, the quantum efficiency, and the light intensity. One or more photochemical reactions are involved in the formation of smog in California and one of the key reactions involves nitrogen dioxide.

Nitrous Oxide Formation

Nitrous oxide, N_2O, the most stable of the nitrogen oxides, is extremely resistant to oxidation to higher nitrogen oxides. It does decompose to nitrogen and oxygen starting at about 600°C[2]. Some nitric oxide, about 10%, is produced from the decomposition if carried out at 1300°[2]. In the laboratory N_2O is produced by the thermal decomposition of NH_4NO_3.

About 0.5 ppm. of N_2O is found in the atmosphere at ground level on the basis of mass spectrographic analysis[3]. Nitrous oxide has also been detected by infrared spectroscopy on the upper atmosphere in about the same concentration[4]. Samples obtained from urban and rural areas showed no significant difference in composition in the mass spectrographic analysis[3]. Furthermore, near infrared measurements from Mount Wilson have indicated uniform mixing in the atmosphere[5]. The rate of change of infrared intensity with solar-zenith angle is inconsistent with the existence of this gas in thin layers in the upper atmosphere[5].

The equilibrium constant for the reaction of nitrogen with ozone indicates that at room temperature almost all of the ozone will be converted to nitrous oxide at equilibrium (Table II). However, the rate of reaction of nitrogen with ozone is quite slow[6].

Hartek and Dondes[6,7] have stated that from their kinetics measurements on the reaction between N_2 and O_3 they conclude that the source of N_2O is the reaction between N_2 and atomic oxygen or ozone in the upper atmosphere. Calculation of free energies and equilibrium constants show that the reaction should occur with N_2O predominating over O_3 except at high temperatures, *i.e.*, 1000°C and above (Table II). At the same time N_2O is destroyed by photolysis in the upper atmosphere. It is claimed that the equilibrium concentration is sufficient to account for the concentration observed.

Since an ozone layer exists at about 25 km., a high rate of production of N_2O in this layer might be expected on the basis of the $N_2 + O_3$ reaction. In this layer, absorption by O_3 should filter out the radiation capable of photolyzing N_2O. Above this layer, N_2O can be produced by the $N_2 + O$ reaction but an appreciable amount of photolysis of N_2O should also occur. Below this layer the concentration of ozone and O drop off appreciably. Consequently, a peak in the N_2O concentration in the O_3 layer might be expected. Experimental results indicate uniform mixing; consequently, the rate of mixing must be about equal to net rate of production.

Although the rates of mixing of nitrous oxide and ozone probably are about the same, the rate of destruction by decomposition outside of the ozone layer is much less for nitrous oxide than for ozone. Consequently, the nitrous oxide, unlike the ozone, is uniformly mixed throughout the atmosphere of the earth.

Goody and Walshaw[8] suggest that bacterial reactions supply N_2O at a rate which compensates for the photochemical destruction in the upper atmosphere and that this is the source of atmospheric N_2O rather than the $N_2 + O_3$ reaction. Some experimental work by Arnold[8] is cited as indicating an appreciable amount of N_2O being produced in the soil. If this occurs, the rate of mixing must be about equal to the rates of bacterial creation and photochemical destruction.

It is reasonable to believe that both the $N_2 + O_3$ reaction and bacterial reaction contribute to N_2O production. Further experimental work appears necessary to determine the relative contribution of each.

TABLE II

Equilibrium Constants for Some Nitrogen Oxide Reactions

Reaction	Kp
$NO + O_3 \rightleftharpoons NO_2 + O_2$	2.29×10^{30} (298°K) 6.93×10^{1} (1500°K)
$NO + \frac{1}{2}O_2 \rightleftharpoons NO_2$	1.17×10^{6} (298°K) 1.07×10^{-2} (1500°K)
$N_2 + O_3 \rightleftharpoons N_2O + O_2$	3.72×10^{34} (298°K) 3.74×10^{6} (1500°K)
$NO_2 \rightleftharpoons NO + O$	1.58×10^{-47} (298°K) 3.82×10^{-4} (1500°K)

Formation of Nitric Oxide from Nitrogen Oxygen

At room temperature an extremely small amount of nitric oxide is formed from the reaction of nitrogen and oxygen both because the equilibrium is so far towards N_2 and O_2 and because the rate of reaction is so slow. However, at the high temperatures resulting from the burning of gasoline in internal combustion engines and of natural gas in furnaces and heaters, appreciable amounts of nitric oxide are produced.

At about 2780°F (1800°K) with 20% oxygen present, 0.44% nitric oxide is present at equilibrium and 90% of the equilibrium value is reached in 12 sec. At 3860°F (2400°K) with 20% oxygen present, 2.00% nitric oxide is formed and 90% of the equilibrium value is reached in about $\frac{1}{2}$ sec. However, Daniels[9] has estimated that to maintain 90% of the nitric oxide formed at 2400°K, it is necessary to cool the gases from 2400°K to 2000°K or below in about 0.02 sec. or at a rate of approximately 20,000°C/sec. At lower temperatures, particularly below 1800°K, the rate of decomposition is negligible. However, the amount of NO formed at 2780°F (1800°K) and even at several hundred degrees lower is not negligible from the standpoint of air pollution.

The adiabatic flame temperatures in gas flames such as occur in domestic and industrial furnaces and heaters are in the range from 3000 to 3600°F. Experimental determinations at UCLA[10] on domestic gas-fired equipment show up to 200 ppm. of nitrogen oxides in flue gases. Even higher concentration of nitrogen oxides would be expected in flue gases from high-temperature industrial furnaces.

Measurements in the exhaust from gasoline, diesel oil and propane-burning vehicles indicate concentrations of nitrogen oxides up to several thousand

[2] Trotman-Dickenson. Gas Kinetics. Academic Pr., New York 71-3 (1955).

[3] R. L. Stobod, and M. E. Krogh. *J. Am. Chem. Soc.* **72**, 1175-7 (1950).

[4] J. H. Shaw, G. B. B. M. Sutherland, and T. W. Wormell. *Phys. Rev.* **74**, 978-9 (1948).

[5] L. Goldberg, and E. A. Muller. *J. Opt. Soc. Amer.* **43**, 1033-6 (1953).

[6] P. Harteck, and S. Dondes. *J. Chem. Phys.* **22**, 758 (1954).

[7] P. Harteck, and S. Dondes. *Phys. Rev.* **95**, 320 (1954).

[8] R. M. Goody, and C. D. Walshaw. *Quart. J. Roy. Meteorol. Soc.* **79**, 496-500 (1953).

[9] N. Gilbert and F. Daniels. *Ind. Eng. Chem.* **40**, 1719-23 (1948).

[10] T. J. Connoly and K. Nobe. Formation of Oxides of Nitrogen in Gas-Fired Heaters. First Rept. on Air Pollution Studies. Rept. 55-27, Dept. of Eng., Univ. of California, Los Angeles.

ppm.[11]. Such concentrations occur during acceleration and cruising, while during idling and deceleration the concentrations may drop to less than 50 ppm[11]. The fact that the nitrogen oxide concentration in automobile exhaust is higher than in flue gas from domestic gas-fired equipment is consistent with the higher flame temperature (4500-5500°F) in internal combustion engines.

Oxidation of Nitric Oxide to Nitrogen Dioxide

Nitric Oxide Oxidation by Oxygen

The oxidation of NO to NO_2 by oxygen goes fairly far over to NO_2 at equilibrium at room temperature, but at high temperatures the equilibrium shifts over towards NO and O_2 (Table II). The rate of reaction is unusual in that it has a negative temperature coefficient[12]. That is, the rate of oxidation is slower at high temperature than at low temperature. Consequently, after nitric oxide is formed by reaction between N_2 and O_2 at high temperatures, relatively little further oxidation would be expected in the homogeneous gas reaction until the gases cooled down appreciably in the exhaust system. At room temperature, for 1000 ppm. of nitric oxide, about 5 min. is necessary to convert ½ of the nitric oxide to nitrogen dioxide. Once the nitric oxide from exhaust gases gets into the atmosphere and is diluted, the rate of oxidation drops rapidly. At 1 ppm. of nitric oxide, it takes about 100 hr. for ½ the nitric oxide to be converted to nitrogen dioxide. At 0.1 ppm. of nitrogen dioxide, it will take about 1000 hr. for ½ of the NO to be oxidized to NO_2.

Analyses of automobile exhaust and flue gas for nitrogen oxides have, for the most part, been for total nitrogen oxides. Much or most of this must be in the form of nitric oxide. However, a few determinations of flue gases from domestic gas-fired equipment by a method specific for nitrogen dioxide indicates an appreciable amount of nitrogen dioxide[10].

Nitric Oxide Oxidation by Ozone

The equilibrium constant for the nitric oxide reaction with ozone is far over towards the formation of nitrogen dioxide both at room temperature and at elevated temperatures (Table II). The rate of reaction of nitric oxide with ozone is extremely rapid even in the ppm. range[13]. At 1 ppm. each of ozone and nitric oxide, ½ of the nitric oxide will be converted to nitrogen dioxide in less than 0.1 min. Even at 0.1 ppm. each of ozone and nitric oxide, 50% conversion takes less than 1 min. If the ozone concentration is of the same order of magnitude as the nitric oxide concentration, nitric oxide will be rapidly converted to nitrogen dioxide. There are other reactions such as NO_2 photolysis which result in the reformation of NO. Also, the ozone can oxidize nitrogen dioxide further to dinitrogen pentoxide. The rate of this reaction is about 10 min. for ½ conversion at 1 ppm. of reactants and 100 min. for ½ conversion at 0.1 ppm. of reactants, so it is 100 times slower than the NO-O_3 reaction. The N_2O_5 formed can then react rapidly with nitric oxide to reform nitrogen dioxide. Thus, a quite complicated cycle of reactions is involved.

TABLE III

Concentrations of N_2O_4 at Various Total Concentrations of NO_2-N_2O_4 Mixture

$N_2O_4 \rightleftharpoons 2\ NO_2$ Kp (25°C) = 0.143 atm.

Total concn. of NO_2-N_2O_4 Mixture (ppm.)	Concn. of N_2O_4 (ppm.)
1000 ppm.	7 ppm.
100	0.07
10	77×10^{-4}
1	7×10^{-6}

Nitrogen Dioxide Photolysis

The nitrogen dioxide produced by the reaction of nitric oxide with ozone and oxygen can be decomposed by ultraviolet radiation. At wave lengths of about 4400 angstroms and below, NO_2 is photolyzed to nitric oxide and atomic oxygen[14, 14a]. Sunlight reaching ground level contains radiation down to 2900 angstrom. Therefore, an appreciable amount of photolysis can occur. The atomic oxygen reacts very rapidly with molecular oxygen to form ozone. It can be seen from the equilibrium constant for the photolysis reaction that very little atomic oxygen is present in thermal equilibrium at room temperature. The reactions heretofore discussed are concerned with thermal equilibrium not involving energy input from an external source. The amount of photolysis occuring depends upon the intensity of the radiation used and the quantum efficiency of the reaction which varies with wavelength. The amount of photolysis of nitrogen dioxide in the atmosphere depends on the amount of sunlight reaching the earth's surface. In the California coastal areas this photolysis is an important key reaction in the complex set of reactions occuring. A significant part of the total ozone concentration appears to result from this reaction.

Nitrogen Dioxide-Dinitrogen Tetroxide Equilibria

In the gaseous state an equilibrium exists between nitrogen dioxide, NO_2, and dinitrogen tetroxide, N_2O_4. The N_2O_4 has a different infrared and ultra violet spectrum than NO_2. Furthermore, the photolysis of N_2O_4 does not begin until about 2800 angstrom unlike NO_2 which absorbs light and dissociates starting at about 4400 angstrom[13]. Consequently, an analysis of the behavior of gaseous nitrogen dioxide must be corrected for the dinitrogen tetroxide in equilibrium with it. This equilibrium is achieved very rapidly; the time for ½ conversion of N_2O_4 to NO_2 being about 10 microsec.[15].

Fortunately, this situation is greatly simplified at lower concentrations. The concentrations of N_2O_4 at various total concentrations of NO_2—N_2O_4 mixtures can readily be calculated (Table III). At a 1000 ppm. concentration of the equilibrium mixture, 7 ppm. are in the form of N_2O_4, while at 1 ppm. of equilibrium mixture only 7×10^{-6} ppm. exist as N_2O_4. Consequently, at concentration of NO_2-N_2O_4 mixtures below 1000 ppm. or 0.1% essentially all of the material is in the form of nitrogen dioxide. As a result, both the spectroscopic determination of nitrogen dioxide and the photolysis mechanism are easier to work with at low concentrations.

Dinitrogen Trioxide Equilibria

A mixture of nitric oxide and nitrogen dioxide will combine to form dinitrogen trioxide, N_2O_3. While the N_2O_3 exists in the liquid and solid states, it is quite unstable in the gaseous state. At 1000 ppm. of equilibrium mixture only 0.1 ppm. of N_2O_3 exists in equilibrium with the NO and NO_2, while at 1 ppm. of equilibrium mixture, only 10^{-7} ppm. of N_2O_3 is present (Table IV). Although the rate at which equilibrium is attained does not appear to have been measured, the rate of reaction between 2 free radical species such as NO and NO_2 should be very rapid. These results indicate that N_2O_3 is not likely to be of significance in any reactions relevant to air pollution.

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[10] See footnote 10, page 98.
[11] M. A. Elliot, G. J. Nebel, and F. G. Rounds. *J. Air Poll. Control Assoc.* **5**, 103-8 (1955).
[12] Footnote 2, 263-5.
[13] H. S. Johnston and H. J. Crosby. *J. Chem. Phys.* **22**, 689-92 (1954).
[14] H. H. Holmes and F. Daniels. *J. Am. Chem. Soc.* **56**, 630-7 (1934).
[14a] T. C. Hall, Jr.; Photochemical Studies of Nitrogen Dioxide and Sulfur Dioxide, Doctoral Thesis, University of California, Los Angeles, 1953.
[15] References 2, 113-15.

TABLE IV

Concentrations of N_2O_3 at Various Total Concentrations of NO-NO_2-N_2O_3 Mixture

$N_2O_3 — NO + NO_2 Kp\ (25°C) = 2.10$ atm.

Total Concn. of NO-NO_2-N_2O_3 Mixture (ppm.)	Concn. of N_2O_4 (ppm.)
1000 ppm.	0.1 ppm.
100 ppm.	10^{-3} ppm.
10 ppm.	10^{-5} ppm.
1 ppm.	10^{-7} ppm.

TABLE V

Equilibrium Constants for Some Nitrogen Oxide Reactions

Reaction	Kp (25°C)
$2\ NO + H_2O + 3/2 \rightleftharpoons 2HNO_3$	2.51×10^{16}
$2\ NO + H_2O + O_3 \rightleftharpoons 2HNO_3$	8.71×10^{41}
$2\ NO_2 + H_2O + \frac{1}{2}\ O_2 \rightleftharpoons 2HNO_3$	1.48×10^{4}
$3\ NO_2 + H_2O \rightleftharpoons 2HNO_3 + NO$	1.09×10^{-2}
$NO + NO_2 + H_2O \rightleftharpoons 2HNO_2$	1.7

Equilibria Involving Nitric Acid and Nitrous Acid

Nitric acid can be formed from the nitrogen oxides by a number of reactions (Table V). These involve the reaction of nitric oxide or nitrogen dioxide with oxygen or ozone in the presence of moisture and the hydrolysis of nitrogen dioxide by water. Nitrous acid is formed by the reaction of nitric oxide plus nitrogen dioxide with water.

The reactions of nitric oxide with water and oxygen or ozone will convert essentially all of the nitric oxide to nitric acid at equilibrium. The reaction of nitrogen dioxide with water and oxygen goes about 98% to completion. That is, about 2% of the nitrogen dioxide remains at equilibrium. Reaction of nitrogen dioxide with water forms about 6% nitric acid at equilibrium, if the concentrations of nitric oxide and nitric acid are the same at equilibrium. If the nitric oxide and nitrogen dioxide are at the same concentration at equilibrium, the nitric acid will amount to about 1.5%. About 20% nitrous acid is formed at equilibrium by the reaction of equal amounts of nitric oxide and nitrogen dioxide with water. All of these reactions are being considered at 25°C.

The final equilibrium mixture will largely be nitric acid. However, since the reaction producing nitrous acid is quite fast[16], initially there would be quite a bit of nitrous acid present in a dark reaction, *i.e.*, in the absence of light. In the presence of light ozone is formed and should react rapidly and completely with nitrogen dioxide and water to form nitric acid. Furthermore, ozone might be expected to rapidly convert the nitrous acid formed to nitric acid, although rate data is not available. Consequently, in the presence of sunlight nitric acid would be expected to form rapidly with the initially-formed nitrous acid probably also being rapidly converted to nitric acid.

Conclusions

The interactions of nitrogen oxides and oxy-acids with each other and with oxygen, ozone, water vapor and ultra violet light result in a complicated set of reactions. Fortunately much can be done to analyze these reactions by the application of chemical thermo-dynamics and kinetics. Note, however, that reactions of nitrogen oxides and oxy-acids with the organic constituents of polluted air are not included. A more complete analysis, which would include even a small number of organic molecules known to exist in polluted air would be considerably more complicated.

The present results indicate that among the nitrogen oxides and oxy-acids; nitric oxide, nitrogen dioxide and nitric acid are expected to be the most prevalent in polluted urban air. Nitrous oxide is also present in urban air but is of less importance because of its lower chemical reactivity. From the standpoint of chemical meteorology, nitrous oxide should be of considerable interest.

The reactions involving dinitrogen pentoxide have not been included in this discussion. Unfortunately, insufficient structural and spectroscopic information is available at the present time to permit the calculations of the thermodynamic properties of N_2O_5.

[16] L. G. Wayne and D. M. Yost. *J. Chem. Phys.* **19**, 41-7 (1951).

Air Pollution from Fluorides*

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The effects of fluorides on vegetation, animals and humans have been under investigation for many years. That they can be damaging to all 3, if present in sufficient quantity, has been well established. There are thousands of references in the literature directly related to this problem. That sufficient concentrations of fluorides may damage living matter and that in some industrial processes large quantities of fluoride may be discharged to the atmosphere has led to the inevitable fact that there is an air pollution problem from fluorides in industry.

These compounds generally produce readily-detectable and identifiable results when they attack plants, animals or humans. In addition, industries potentially capable of producing large quantities of fluoride effluent are often relatively isolated and located in agricultural areas. This makes it a relatively simple matter to identify an industrial plant which may be producing such injury. Also from an economic standpoint losses can be high. Whether or not an economic loss occurs depends upon 2 factors: first, whether or not there is sufficient contaminant to produce damage and, second, whether or not there is anything to be damaged. Thus the nature of the environment in which an industrial plant is located will determine to a large degree whether or not a serious fluoride pollution problem will exist. In heavily industrialized areas where a producer of fluoride pollution is but one of many offenders it may be difficult to place the blame for economic loss. In the case of a single plant located in the middle of an agricultural or dairy area, however,

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the finger can be pointed with ease in the event of damage to crops or animals.

There has been a long history in this country of severe crop and cattle damage in several areas caused by air pollution from fluorides. In spite of an extensive knowledge of both the cause and the effects of fluoride pollution, the problem is becoming more and more severe. There are at least 2 basic reasons for this. First, in those industries which have a history of this problem, there has been a vast increase in production and a simultaneous increase in the quantities of fluoride by-products produced.

In some instances, development of new processes has increased fluoride production. Second, the introduction of industries known to have fluoride pollution problems into new states or new areas where little is known of the problem has intensified it.

There are 2 major sources of fluorides responsible for the pollution problem. These are rock phosphate and fluorspar and other ores of fluorine compounds. A brief review of production figures for these materials in recent years will define the extent of the problem.

Phosphate Rock

Phosphate rock is utilized primarily for production of superphosphates (fertilizers), food and medicinal phosphates, elemental phosphorus, phosphoric acid, ferro-phosphorus, stock and poultry feed and it is applied directly to the soil as fertilizer. The important sources are in Florida, Tennessee, Idaho, Montana, Utah and Wyoming. Production has increased rapidly during the last 10 yr. as indicated in Table I.

Note that while production in Tennessee has remained relatively stable, it has increased substantially in Florida and the Western states.

Utilization of this rock is important in understanding the impact on fluoride pollution. In general, fertilizers (superphosphate) are processed near the source of the ore, while availability and cost of power has a greater influence on the location of industrial plants manufacturing elemental phosphorus.

Table II, giving the uses of rock phosphate from the 1950 figures of the Bureau of Mines Mineral Yearbook (1953), illustrates this fact.

TABLE I
Production of Phosphate Rock (Long Tons)
(from Minerals Yearbook—U. S. Bureau of Mines)

	1945	1950	1952
Florida	3,814,935	8,597,227	9,205,138
Tennessee	1,260,849	1,472,017	1,444,737
West. States	323,955	1,044,915	1,381,338
Total U.S.	5,399,739	11,114,159	12,031,213

TABLE II
Utilization of Rock Phosphate
(Expressed in %)

	Florida	Tennessee	Western States
Superphosphates	68	22	26
Phosphates, Phosphoric and Elemental Phosphorus	5	60	38
Direct application to soil	7	16	9
Other uses	20	2	27

Thus Florida, which does not have sufficient power for phosphorus production uses most of the rock phosphate mined for agricultural purposes, while in Tennessee and in the Western States, phosphorus is the predominant product because of abundant power.

Rock phosphate is an important source of fluoride pollution because high grade ore contains 2 to 3% F. The phosphorus is present as fluorapatite and thus there is a fixed ratio of fluoride to phosphorus. This means there are roughly 25 tons of F available/1000 tons of rock processed. In those cases where the rock is processed in such a way that the fluorphosphate is broken down, large quantities of fluorides can be released.

In production of *elemental phosphorus*, fluoride may be released in several of the steps in production. Because of this complexity, it is important in phosphorus production operations to undertake fluoride balance studies to determine where fluoride losses are occurring before an intelligent approach can be made to controlling air pollution. It is also valuable from an economic standpoint to be able to apply control measures only where they are needed.

Some of the fluoride loss occurs in the first step of calcining or sintering the ore. The amount depends primarily on temperatures used. Significant losses may occur in the electric furnaces. Part of this escapes directly into plant air and then to the atmosphere, while the remainder is in the offgas which is usually burned with subsequent release of fluoride to the atmosphere. Substantial amounts of fluoride may be trapped in the furnace slag, but part of this may be released to the atmosphere during cooling in slag pits. Distribution of fluorides among these various sources will vary with variations in the process. Thus the necessity for study where these losses occur is obvious.

The important fact to bear in mind is that for every ton of rock which is processed, 40 to 60 lb. of F may be released. In a plant processing 1000 tons of rock/day this can produce a serious air pollution problem if uncontrolled. Manufacturers of elemental phosphorus can confirm this.

In production of *superphosphate*, the rock is treated with sulfuric acid with resultant release of fluoride as HF and silicon tetrafluoride. This is an operation involving handling of large tonnages of ore and thus large quantities of gaseous fluoride. By the very nature of the product, fertilizer manufacturing plants are generally located in agricultural areas and thus may produce damage.

A new development in this industry has been concentrated superphosphate, made by producing phosphoric acid from the rock, and treating additional rock with this phosphoric acid. This has resulted in an even greater release of fluorides. In some areas it has changed the situation from borderline to definite damage.

In 2 areas, one in Tennessee and one in Florida, the situation has been complicated by the operation of many plants (10-12) producing superphosphate, phosphoric acid, animal and poultry feeds and elemental phosphorus, in a relatively small area (10 mile radius). The joint contributions of all the operations has produced a pollution problem of major proportions. In such a situation assessing the blame for damage is difficult. There are wide variations in process, size of operation, control measures, *etc.*, among the plants. In addition, since the ultimate criterion is the total amount of fluoride emitted in an area, the permissible effluent/plant must be reduced substantially. It is absolutely necessary that such groups of industries work closely together in matters of evaluation and control if the situation is to be kept under control.

Fluorspar and Other Fluoride Ores

The utilization of fluorides, particularly in production of aluminum and steel provides another large-scale source of fluoride pollution. From 1948 to 1952 the consumption of fluorides by industry increased from 406,269 to 520,197 short tons. This is exclusive of cryolite, both natural and synthetic. Of the fluorspar consumed, the steel industry used 61% in 1948 and 53% in 1952; hydrofluoric acid increased from 24% in 1948 to 34% in 1952, other uses (chiefly glass and enamel) decreased from 15% to 13%. The steel industry was a major user of fluorspar, primarily as a flux in

the manufacture of basic open hearth and basic electric steel. It is difficult to assss the proportion of fluorides released to the atmosphere, but the industry has had its problems.

The Aluminum Industry

In the production of aluminum metal from the refined ore (alumina) the source of fluoride is also incidental to the process. This is an operation in which alumina is mixed with fluorides in electrolytic furnaces. The fluorides act as a flux and are released to the atmosphere as a result of decomposition under high temperature conditions. There are 2 basically different kinds of furnaces used, one of which employs pre-baked electrodes, the other provides for baking of the electrodes from the heat of the furnace during operations. In both instances the fluorides are carried upward by convection and in the absence of local exhaust, are generally released to the atmosphere through roof louvers. All these furnaces operate continuously. The quantities of fluorides produced vary with the number and sizes of the furnaces and operating procedures. The quantity of fluoride escaping to the outside depends not only upon these factors but also on whether or not attempts are made to control the effluent. There have been 2 major approaches to control. One of these is the installation of sprays and roof monitors, permitting the fluorides to escape from the furnaces and attempting to trap them at roof level. This has not produced satisfactory results. A second method of control is to provide local exhaust ventilation on each furnace and to pass the exhausted air through scrubbing towers in an attempt to remove the fluorides at this point. The effectiveness of this system depends primarily on the kinds of hoods used, the volumes of air handled and on the effectiveness of the scrubber system. Such control systems are extremely costly because of the large numbers of furnaces involved and consequently the very large amounts of air which may be handled. In some of these plants 2½ to 3 million ft.^{3} of air/min. are being exhausted and scrubbed.

Aluminum production depends primarily on ore and power. Plants are thus located either near the source of ore or in areas of low power cost. There are at the present time 6 aluminum producers operating in the states of Washington, Oregon, Montana, Texas, Arkansas, Alabama, Louisiana, North Carolina and New York.

The Pacific Northwest has had a long history of damage claims as a result of injury to cattle and vegetation. Obviously, aluminum reduction plants cannot operate without controlling the fluoride effluent. The severity of the problem depends on the amount of emission, climate, meteorology, and the nature of the surrounding territory. Setting permissible levels for the amount of fluoride effluent is difficult. The gaseous fluorides may be retained and built up in vegetation, so that time is a factor. The most satisfactory method found for evaluation of a fluoride pollution problem is a continuing vegetation sampling program. A grid system must be set up and periodic samples of different kinds of vegetation collected periodically during the growing season. This will give the best measure of the effectiveness of control measures.

In spite of tremendous amounts of work done in investigating this problem there are few valid criteria for fluorides in vegetation. There are 2 problems: (1) is related to the amount of fluoride in the vegetation which will cause damage and (2) is the amount of fluoride in vegetation which will produce injury in animals which consume it. Both are complex. The levels which will damage vegetation vary with species over an extreme range. Fruit trees and some types of flowers are particularly susceptible to injury at low concentrations. Levels in grasses and animal feed which will injure cattle also vary with the condition of the cattle, fluoride intake from water and other sources and time of exposure.

TABLE III
Fluorides in Vegetation (Ppm.)

Product	Distance in Miles: 0 to ½	1	2	4	6	
Fertilizer (with collection)	3-21	6-14	0-15	3-24	6-21	No complaints
Fertilizer (partial control)	81-153	39-120	16-81	14-47	6-44	No complaints
Fertilizer (partial control)	52-285	35-177	19-41	9-48	6-17	No complaints
Fertilizer (partial control)	130-845	78-327	100-223	64-114	—	Claims
Phosphorus (Averages—several plants)	62-495	20-316	26-212	8-140	12-110	Claims
Aluminum reduction (Averages—several plants)	160-1600	95-1000	36-380	8-135	4-87	Claims

Typical Examples

Because of the wide variety of conditions, both as to amounts of effluent, its distribution, and the type of area being polluted, it is difficult to give average data. A few examples may serve to illustrate the kinds of contamination involved. These are shown in Table III.

These figures represent the kinds of values which can be built up in vegetation as a result of the operations discussed. They are a brief summary of thousands of values. Levels will vary with the many factors discussed above. Extensive damage has occurred and in many instances the cost of claims has been staggering.

With the volume of data available, obviously no plant producing large amounts of fluoride effluent can afford to neglect the air pollution problem. Vegetation levels should be kept below 30 ppm. in order to prevent damage to cattle. Ventilation and collection should be installed at the time such a plant is constructed, for without such control, trouble will develop. Adding such installations after operations have begun will increase their cost substantially. The cost of fluoride damage throughout the country to date has been many millions of dollars. If additional millions are not to be lost, dissemination of fluorides must be substantially reduced. The question cannot be resolved by lawyers or public relations specialists. It can be handled only by sound engineering.

Public Health Responsibilities in Air Pollution*

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New Jersey State Commissioner of Health

Air Pollution is the general term applied to contaminating substances in the atmosphere. These substances may exist in the form of dusts, vapors, gases, mists or odors and are derived from multiple sources. The chief sources in urban and industrial areas are households, public and government-owned installations, and industrial plants. In Los Angeles, figures have been quoted that 40% of air pollution is the responsibility of industry, 60% due to other community activities. But it may be stated with assurance, that much physiologically significant air pollution stems from industry.

Official agencies, research institutes, laboratories, universities and large industrial corporations are all increasingly concerned with the complex problem of sources of air pollution and their control. It should not be forgotten that the inauguration of air pollution control is primarily for the benefit of human beings.

Health and Air Pollution Defined

Basically, what do we mean by health? Until fairly recently, a person was considered healthy who was not sick. The World Health Organization defines health as follows: "A condition of complete physical, mental and social well-being, and not merely the absence of disease or infirmity." This is a descriptive concept; not a yardstick for quantitative or qualitative measurement. By this concept, therefore, an atmospheric pollutant is any physical, chemical, or parasitic agent to be found in our natural or artificial environment capable of being air-borne and producing human death, illness, disability, injury or discomfort.

The Air Pollution Conrtol Act (1954) adopted by the State of New Jersey further extends this definition by defining air pollution as "the presence in the outdoor atmosphere of substances in quantities which are injurious to human, plant, or animal life or to property, or unreasonably interfere with the comfortable enjoyment of life and property."

A health problem becomes a public health responsibility if or when it is of such character or extent as to be amenable to solution only through systematized social action. Its relative importance varies with the hazard to the population exposed. This hazard may be qualitative, in terms of disability or death; quantitative, in terms of proportion of population affected; it may be actual or potential. [1]

The primary responsibility of the public health administrator is the welfare of the total community under his jurisdiction. In a state as highly industrialized as New Jersey, a public health administrator, while not an engineer, a meteorologist, or a micro-analytical chemist, finds one of his duties is the investigation and solution of the air pollution problem.

Health Factors

The health of any community is a relative function and is influenced by many physical, pyschological, social and economic factors, including heredity, race, sex, age, income, nutrition, water and food sanitation, medical care, pathogenic micro-organisms, man-made air contaminants, occupation, housing, recreational facilities, climate and pollens. Adverse effects on health can be defined in a variety of terms. The relative public health significance of atmospheric pollutants depends upon their potency to produce, directly or indirectly, death, illness, disability, injury or discomfort. Public health and medico-legal problems created by some atmospheric pollutants, may require much time and research to solve.

In this paper the word *injury*, as distinguished from death, illness or disability, includes physiological responses of such degree that the affected person becomes conscious of the reaction, that an informed observer can detect the reaction, that the majority of persons similarly reacting would find the given physiological response a definite annoyance, and finally, that at least 2% of humans exposed to the given pollutant or combination of pollutants under standard conditions would respond in an equal or more severe manner.

Action Can Produce Relief

Some persons not trained in medicine, who have never been responsible for diagnosis and treatment of such sufferers, say that sneezing, coughing and swelling, congestion and watery discharge from mucous membranes, *etc.*, are but normal physiological reactions designed to dilute and wash away the irritating substance. It is true that such physiological reactions are normal responses to irritating substances, but this does not prove that the irritating substance is normal, desirable or necessary. Thousands of hayfever sufferers, for example, cannot be expected to accept their plight under the title *normal physiological response* when they learn that a sound weed control program can markedly reduce atmospheric pollution by the offending pollens, and thus give them great relief from their undesired and unnecessary physiological responses.

An occasional citizen responds allergically to tomato and may develop an asthmatic response or have hives with severe itching. These reactions may require medical care and may prevent the individual from pursuing his usual activities or earning his livelihood. At present, each person eats tomatoes or strawberries, *etc.*, at his own peril. This is not because the reaction is medically explicable by reasonably well understood physiological and immunological reactions. It is because only a very small percentage of persons has such a response to these foods. However, if a high per-

* Excerpts from an address presented at the 1st Air Sanitation Seminar of the Mid-Atlantic States Section of APCA at Rutgers University, April 13, 1955.

[1] H. S. Mustard. Introduction to Public Health.

centage of the citizens reacted in this manner to tomato or strawberry, it would become mandatory to indicate on every menu which foods or desserts contained these food items even in traces and in non-recognizable form such as in juices, icings, *etc.* This distinction may be applied in the field of atmospheric pollution.

Clearly, from the public health administrator's point of view, there are many known harmful atmospheric pollutants, including chlorine gas, hydrogen fluoride, sulfur trioxide, lead fume, beryllium, silica dust, anthrax spores, pollens, radioactive dusts, *etc.* Furthermore, there are many more such as chromates, nickel carbonyl, aromatic amines, *etc.*, which are strongly suspect for increasingly good medical and epidemiological reasons. Time and research will eliminate the suspicion or add these to the list of known substances capable of producing harm to humans via the atmosphere . . .

Conclusion

Legislation should be of the performance rather than of the punitive type, although a penalty clause is always necessary in the event of a recalcitrant offender. Despite the lack of absolute standards and full knowledge as to all the causes of air pollution, it is possible to write codes or regulations broad enough in scope, and flexible enough to allow for scientific discoveries concerning the elimination of air contaminants.

With improved public relations, and self-regulation by industry, with education and better understanding by local citizens, and with impartial and non-discriminatory legislation and administration by the State, the problem of atmospheric pollution will yield to diagnosis and treatment.

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The Disposal of Coal Refuse

Technical Coordinating Committee T-4 Coal Report

Spontaneous heating within a refuse pile, caused by oxidation of the coal, absorption of moisture, and oxidation of pyrite, will result in a fire if air flow through the pile is within a certain critical range. Outside of range at the lower end is the well compacted pile (little air movement) and at the upper end is the pile of large, non-friable coal (free air movement). It has been observed that both types of piles seldom catch fire.

However, fires are of general occurrence in bituminous refuse piles because their construction is such that the air flow falls within the critical range. This construction is a normal characteristic of aerial tram, incline, larry, and careless truck-disposal operations. As such piles become higher, size segregation, void space between larger pieces, erosion cuts, settlement, and the development of slippage fissures enable restricted upward circulation of air. This chimney effect within the pile promotes spontaneous heating and utimate combustion of the refuse pile.

Many coal operators have found that by observing proper care in the selection of dump sites and the methods of disposal, the occurrence of refuse fires can be eliminated. Such practices in pile construction as compaction by roller in thin layers, crushing the larger pieces to an intermediate size, reducing the pile slope, and the use of a terraced face will reduce spontaneous ignition and permit control of fires in unsealed piles.

For even better protection against refuse pile fires, some operators have sealed the piles with clay or fine refuse. This procedure is very suitable for hillside sites because the hillside reduces the exposed edge area and clay is available for the seal at the rising pile level.

Methods have been developed whereby existing refuse-pile fires can be extinguished. Water can be used to cool areas to be extinguished, but the use of water to stop the fire has not been successful. Satisfactory results have been obtained by sealing the burning pile with compacted refuse, cleaning plant sludge, or refuse fines.

Introduction

For many years the prevention of fires was given little consideration in the disposal of coal refuse. Fires were prevalent and atmospheric pollution (frequently intensified in valleys by atmospheric conditions) was severe in mining communities. And the trend to mechanized and full seam operations, resulting in increasing amounts of refuse for surface disposal, aggravated the problem. Atmospheric pollution became a problem of serious concern to all coal operators.

Articles on the mining and preparation of coal usually contain little or no information on refuse disposal, and the subject is rarely treated comprehensively as a matter of importance in its own right. Lack of interest in the subject is clearly manifested in the technical literature, and it seems fair to say that until recently both the industry and the public were indifferent to burning gob piles. By no means confined to the coal industry, however, an attitude of indifference to atmospheric pollution from all sources, domestic and industrial, generally prevailed in most heavily-populated centers. In fact, smoke was regarded as visible evidence of good times.

In recent years the public attitude has changed. Increasing demand for abatement of atmospheric pollution is a factor which must seriously be considered by an operator who intends to construct a new refuse pile. The construction of a new pile should be undertaken with the realization that, unless properly constructed, the pile will be a potential menace for many years after operations in the mine and supervision of the pile shall have ceased. The compacting, sealing and crushing procedures herein recommended are designed to prevent or minimize refuse fires. The cost of these procedures is not excessive and the premium may be regarded as a contribution to community development and good public relations and as insurance against liabilities and costs in the control of refuse fires. In fact, some operators have reduced disposal cost, particularly in maintenance, depreciation, and even loss of trucks by changing from the driver's choice method of disposal to compacted layer construction of refuse piles.

Layer disposal and compaction of refuse has reduced greatly the incidence of spontaneous ignition; such construction is easily achieved by the use of trucks for refuse disposal. Trucks have largely displaced aerial trams and larries in moving refuse from the cleaning plant to the dump because they are more flexible in operation and permit dumping and compacting in layers of desired thickness over an area of any configuration. The incidence of spontaneous fires could be still further reduced, possibly eliminated, if the exposed faces as well as the top of refuse piles were intensively compacted. The development of procedures for overall compaction is necessary for reasonable fire protection in unsealed refuse piles.

Practice in the compaction of refuse varies greatly, especially with respect to layer thickness, equipment for compaction and procedure used at the layer edges. Layer thickness, although fairly constant at each pile, has been observed generally to range at different operations from 1 to about 10 ft. Compaction may be effected by bulldozer and directed travel of trucks (the weight of which varies greatly) or additional compacting equipment attached to the bulldozer may

be used; and procedure at the layer edges (pile faces) varies from compaction with a bulldozer (or bulldozer and roller) on low slope piles to complete disregard of compaction on high slope piles. Edge compaction presents little difficulty on the low slope piles which can be constructed in flat or rolling land, but it is a difficult problem on the high slope piles which frequently must be constructed in very hilly or mountainous terrain.

Therefore, despite its advantages, layer disposal of refuse, as usually practiced, does not assure freedom from spontaneous ignition. Frequently, the layers are so thick that rollers and trucks can compact only a relatively small portion (the crust of each layer) and quite generally the edges of the layers are not compacted at all. Furthermore, when layers are leveled it is common practice to push refuse over the edges. Therefore, in most steep piles there is substantial edge area of uncompacted refuse and increasingly an accumulation of segregated refuse along the faces, the fines along the upper slope and the coarse sizes rolling to the base of the pile. The danger of sponstaneous ignition increases with the height and slope of such loosely constructed piles; and, of course, the danger of fires from irresponsible persons is always present.

Disposal and pile construction procedures should be designed to prevent or minimize spontaneous, incidential or deliberate ignition of the refuse; in case of fire, to permit access to the pile for control and extinguishment of the fire; and at the termination of the operation, to facilitate the eventual establishment of vegetation. This definition goes beyond the prevailing concept of responsibility in the matter; but it is a practical, attainable goal and the coal industry is morally bound to its acceptance. Coal will be mined in this country for many centuries and each generation is obligated to dispose of its refuse in such manner that it will be neither a potential source of atmospheric pollution nor a scar on the landscape. This goal can be attained only by the interest of top management in the problem. Interest at the highest level is quickly reflected by key personnel at the plant and results in general appreciation and observance of improved methods of pile construction and maintenance.

Spontaneous Heating of Coal and Coal Refuse

Spontaneous heating of coal and coal refuse piles is essentially an oxidation phenomenon, involving coal and associated pyrite and impure coal substances, but it is recognized that moisture vapor contributes in varying degrees to the self-heating process. There are 3 major heat producing reactions in this natural process.

Oxidation of Coal

Several investigators have attributed spontaneous heating to the formation of a coal-oxygen complex in which the oxygen is in an active condition, as in a peroxide formation. Jones and Townend, in particular, have compared coals and carbons with oils, rubbers and foodstuffs in respect to the mechanics of oxidation (peroxide effect) and they have measured the formation, decay and stability of peroxygen in coals and carbons:

> The presence of adsorbed water is a necessary condition for the formation of this complex; this is more especially true of carbons . . . On coal exposed to air at ordinary temperatures the peroxygen complex builds up to a maximum concentration and then decays. The overall oxidation proceeds without interruption (or even at an accelerated rate) even when the peroxygen concentration has become very small . . . The nature of the overall oxidation process (probably of the breakdown process) is greatly different above and below the critical temperature range of 70 - 80°C. Below this range, CO is the main gaseous product and very little CO_2 is formed; above it, CO_2 and water are formed in much greater proportion than CO.

Moisture Absorption

Many observers have noted a temperature rise in coal piles when dry coal is piled over a wet area or against wet coal, when rain follows prolonged dry spells, *etc.* Berkowitz and Schein have reported temperature rises from 1.1 to 36.1°C. (at rates from 0.30 to 6.70°C./min.) in calorimeter tests on air dried Commonwealth coals when oxygen and nitrogen saturated with moisture vapor were passed through the coals at 25° and 40°C. Only insignificant temperature rises were observed with dry oxygen.

> . . . In recognizing that the heat of wetting may act as an *important trigger* it is not, however, essential to suppose that it can only affect initially dry coal. While the heat of wetting/unit gain of water will necessarily decrease as the initial moisture content of the coal increases, it can readily be shown that it remains large enough to influence the oxidation velocity of any coal whose moisture content has been depressed below its normal equilibrium value by a succession of warm dry days. The only proviso is that the coal shall be a reactive one in the sense of possessing a large internal surface. With unreactive coals, such as coking coals, even the maximum heat of wetting will normally be too small to affect the oxidation rate appreciably.

Coal has the physical structure of a gel (porous structure and large internal surface area) and it is generally considered that the spontaneous heating of coal, as determined by the above oxidation and moisture absorption reactions, is a function of surface area, of which the internal area in a substance of such wide variation in porosity, is the determining factor. However, Malherbe observed little correlation between oxygen absorption tests and surface area measurements on South African coals and concluded that if the oxygen absorption test is a true measure of the tendency to spontaneous ignition then this tendency is not primarily dependent upon the extent of the internal surface but is influenced more by the chemical properties of the surface. Quite likely, both physical and chemical structure determine the extent of spontaneous heating.

Oxidation of Pyrite

The atmospheric oxidation of pyrite is a factor of importance in the spontaneous heating of coal and coal refuse, directly because the reaction is strongly exothermic and indirectly because associated coal is shattered by the reaction and the active coal area is increased. Obviously, the more finely-dispersed in coal and carbonaceous substances, the more effective is the pyrite in this respect. Also, pyrite is the major cause of stream pollution in coal mining districts. It has been estimated that annually about 1,000,000 tons of sulphuric acid is discharged from coal mines into the streams of Pennsylvania. The dry and wet oxidations proceed as follows; atmospheric moisture vapor favoring the more strongly exothermic wet reaction, with formation of sulphuric acid:

$$FeS_2 + 3O_2 \longrightarrow FeSO_4 + SO_2$$

$$2FeS_2 + 2H_2 + 7O_2 \longrightarrow 2FeSO_4 + 2H_2SO_4$$

GENERAL CONSTRUCTION OF REFUSE PILES

Pile Location

In hilly and mountainous mining districts, coal refuse piles are generally (usually necessarily) constructed on hillsides. The placement of piles has been determined largely by economic factors, such as land values and space requirements in the vicinity of the cleaning plant and the cost of moving refuse from plant to pile (the largest element of cost in its disposal). The topography of the disposal site in relation to refuse capacity and pile construction costs (especiallly in sealed piles) is a consideration of increasing importance.

Disposal Area

Sufficient land for the disposal of refuse during the mine operation expectancy should be acquired within reason-

able transportation distance from the preparation plant. If disposal space runs short, it is common experience that adjoining land values suddenly increase.

Preparation of Site

The site of the pile should be cleared of vegetation. The importance of this precaution is generally recognized, but it is pertinent to observe that occasionally piles are allowed to encroach upon grassy or even wooded land bordering the site.

If a pile must be located on a hillside without appreciable level land at the base, it may slide. Possibility of this occurrence may be reduced by digging a deep wide trench across the hill at the base of the pile, throwing the earth forward to form a wall in front of the trench and filling the trench with refuse.

Disposal Procedures

Compacted layer construction is the common feature of all safe disposal methods. In addition, for greater protection the pile faces should be sealed or they should be sloped or terraced to permit compaction and control of possible fires. Also, in limited but successful use is the procedure of crushing the refuse to a fine size consist.

Drainage

Excessive water run-off from a hill back of the pile should be diverted from the pile. This will reduce erosion at the faces and operating difficulties during heavy rains (especially on heavy clay refuse).

Mine Rock

Since uncrushed mine rock cannot be compacted until it disintegrates, it will form large void spaces in a pile and favor the access of air, particularly if it is dumped near the edges of a pile. The disposal of rock at the edges of a pile is an open invitation to fire. It is advisable therefore to crush rock for disposal with plant refuse.

Extraneous Refuse

All extraneous mine, household and neighborhood refuse, such as mine timber, boxes, paper, garbage, bottles and cans should be excluded from piles. Combustible or incombustible, all extraneous refuse may accelerate spontaneous heating of the coal refuse. Curiously, it is usually dumped where it is most certain to cause trouble—along the edges of a pile.

Unsealed Refuse Piles

Thinner layers, intensive compaction and reduced or terraced slope to permit access to the faces are features of pile construction which will reduce spontaneous ignition and permit control of fires from any cause in unsealed piles. The goal should be the development of acceptable methods of safely storing unsealed coal refuse against the hazards of spontaneous, incidental and deliberate ignition. At present, at least, for most refuse piles and procedures in use, that degree of security is not assured.

Thin Layers

At one typical operation refuse is dumped from a scraper (loaded weight over 50 tons) in layers about 12 in. thick over the top of the pile except at the edges (considerable crushing being effected in this manner) and then it is pushed to the edge with a bulldozer. The faces slope at the natural angle of repose but hot spots develop only infrequently. The interior, main body of the pile is so well compacted that it would not burn. Thin layer dumping with this type of equipment is strongly recommended; in conjunction with lower slope or terraced construction to permit access to pile faces, it is the safest procedure for the construction of unsealed piles.

Layer thickness is the primary factor in good compaction of refuse, just as it is in the compaction of earth for roads, fills, dams, *etc.* The effects of compaction decrease with distance from the layer surface and are rarely pronounced at a depth of 2 ft. Thick layers are honeycombed with void spaces which support combustion. For this reason, it is recommended that unsealed refuse should be deposited in layers not exceeding 2 ft. in thickness and that in all refuse operations, transport equipment should be used which is capable of dumping refuse in layers or in piles of such height as can be easily leveled to the desired layer thickness. In the purchase of equipment, this consideration should be the dominating factor.

Compaction of Refuse

In layer disposal of refuse by trucks, compaction is frequently effected only by the vehicles, and such compaction is usually superficial (especially at the edges) and it is poorly supervised, largely a matter of driver's choice. In conjunction with truck compaction, it is recommended that refuse be compacted with a dozer and a heavy roller, such as sheepsfoot roller, a smooth cylindrical roller (easily made in the shop) or a pneumatic tire roller.

At 2 mines of a mid-western operator, proper compaction methods have been successfully used. The refuse is deposited on the pile by trucks of 5 ton capacity. The type of refuse obtained from these 2 mines is such that a considerable portion of it will slack when exposed to the atmosphere. Considerable disintegration takes place within 24 hr. At these 2 mines, the 5-ton loads after slackening are bulldozed. Then a drag composed of four 60 lb. railroad rails is attached to the bulldozer and the pile is further leveled. Finally, a roller consisting of 2 concrete cylinders 5 ft. in diam. by 4 ft. long is attached to the bulldozer for the final compaction. It is interesting to note that the drag and the roller may be passed over the pile 3 or 4 times.

One coal company uses a concrete filled homemade roller (diam. 4 ft. and length 6 ft.) at their mine. Five x $\frac{3}{8}$ in. cone refuse is put down in layers about 5 ft. thick and is compacted with the roller. Located at the head of a valley, most of the pile is naturally sealed by the hillside and the lower end is sealed by a heavy clay wall which extends across the valley.

And of particular interest are 2 rollers recently developed by another company, *viz.*, a dual wheel semi-trailer for use on pile tops and slopes and a side roller which is described further in this section. Both are equipped with pneumatic tires which (as in the case of trucks) have proved to be excellent compacting agents. For intensive compaction, they are heavily loaded with steel scrap. The several methods of compaction are shown in Fig. 1.

Incidentally, one operator found a diesel powered roller to be unsuitable for use on heavy clay refuse—during hard rains the roller stalled in slushy refuse and needed assistance from a bulldozer for further locomotion.

Weathered Refuse

It is advisable to permit truck heaps of refuse to weather as long as possible before they are leveled into layers of the desired thickness and compacted. The longest possible weathering period at the edges before compaction is particularly desirable. Exposure to the atmosphere permits the initial oxidation of fresh, reactive coal and pyrite to proceed rapidly under conditions in which the heat is dissipated; and it causes disintegration of slate and rock in the refuse so that subsequent crushing and compaction by truck or roller is greatly facilitated. The speed of weathering or disintegration is

greater in some districts than in others but it is a factor which always should be utilized to the utmost advantage. The larger the pile, the longer will be the weathering period for each layer and the more advantage can be derived from the weathering process. Some operators consider this factor to be of such importance that they have 2 refuse piles, one of which is weathering while the other is receiving refuse.

Coarsely Crushed Refuse

If trouble is experienced in compacting mine refuse, pickings and large sizes of washery refuse, much benefit will be obtained by crushing to an intermediate size, 3 in. or less, before disposal in the usual manner.

Reduced Slope

Reduction of the slope is the simplest way to provide access to the pile sides for compaction and assure control of fires. It is possible that the bulldozer or the dozer and trailer previously mentioned can be used effectively to control hot spots on weathered refuse with slopes as high as 50%. Size consist, weathering characteristics and crushing effect of the compacting equipment will determine the facility with which the bulldozer can operate on steep slopes. If difficulty is encountered, it is suggested that the refuse be deposited in thin layers well within the edge and that it be weathered and crushed with a roller before it is pushed to the edge with the bulldozer; or better, crush the edge refuse before it is dumped. In this manner the slope can be better compacted in depth.

Generally, reduction of the slope from the normal angle of repose to a maximum of about 50%, as recommended, does not entail serious loss of pile capacity, as may be noted in Fig. 2. OA is a 100% slope line (roughly the normal angle of repose) and OB is the recommended slope. Hillside slopes of 10, 20 and 30% through points of origin on the base line may be seen. Comparison of the slope lines permits a qualitative estimation of the effect of pile slope, hillside slope, pile height, and level land (apron) in front of the hill on the capacity of the pile. Except on very steep hillsides with no level land on the pile base, the pile capacity is not seriously affected by the slope reduction to about 50%.

Terraced Side

Access to the pile side can also be provided by terraced construction. The terrace also reduces the fire hazard by checking the draft along the slope. The use of some type of *side roller,* (Fig. 1), is of particular interest for use on terraced piles with slopes which are inaccessible to conventional compacting equipment. The roller consists essentially of heavy dual wheels with pneumatic tires closely mounted on a short axle which is connected by a long frame to the side of a bulldozer. An adjustable jack on the axle and pin couplings at the dozer permit vertical adjustment of the roller to the contour of the edge of the pile. A box loaded with steel rails is mounted on the axle near the wheels. The loaded weight of the roller is about 10 tons. When the dozer is operated along the edge of the pile, the roller extends over the edge and gives a rounded and compacted edge. The slope can be rolled for a distance of 12 ft. below or above the level of the dozer. The entire edge area of each layer or bench can be quickly compacted if the side roller and semi-trailer previously mentioned are both attached to the dozer.

Sealed Refuse Piles

For better protection against fires, some operators seal refuse piles with clay. At one operation on the Allegheny River, layers of clay about 15 in. thick are interposed between compacted layers of refuse about 36 in. thick and the layer edges also are sealed. In general, however, clay is used only at the layer edges; in conjunction with compacted layer disposal of the refuse this practice has eliminated fires at several mines. Sealed construction offers maximum protection against fires, pile concealment and protection of pure water sources.

Hillside sites are favored for the construction of sealed piles because the hillside reduces the exposed edge area and clay is available for the seal at the rising pile level. Narrow valleys with steep hillside slope restrict the pile width and capacity and entail high pile construction costs; and particularly in narrow valleys, provision for drainage of water along the valley channel in front of the pile seriously reduces the available disposal area. Rolling terrain and broad valleys with low hillside slope, however, permit piles of substantial width; and in such locations pile capacity is high and compacting and sealing costs are low. Lowest sealing costs can be achieved in valleys which can be filled with refuse from one hillside to the other, across the span of the valley, as in a *dry valley* or a valley in which the drainage above the pile is not excessive and can be handled

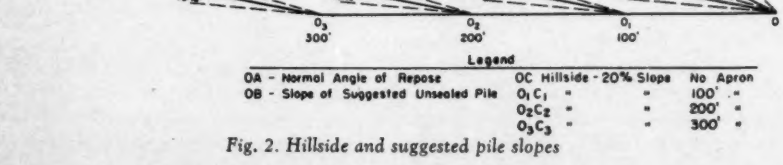


Fig. 2. Hillside and suggested pile slopes

A. Dual wheel semi-trailer at Renton.

B. Dumping refuse with a scraper in layers about 12 in. thick at Robena (very good compaction.)

C. Side roller at Ocean

D. Concrete filled home-made roller at Arkwright

Fig. 1.

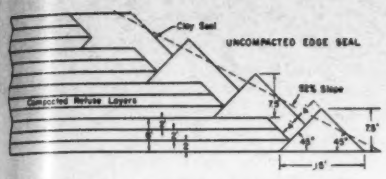


Fig. 3. Uncompacted edge seal

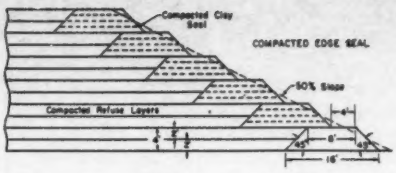


Fig. 4. Compacted edge seal

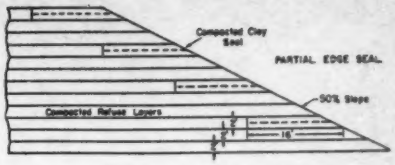


Fig. 5. Partial edge seal

in a concrete pipe on the valley floor under the pile (the pipe being heavily covered with clay to prevent access of air to the interior of the pile, possible fire and corrosion of the pipe). In such locations, if the upper end extends to the head of the valley or feathers out against the valley floor, only a seal across the valley at the lower end of the pile is necessary.

Strip operations afford an opportunity for the complete sealing of refuse. In operations on clean streams in Pennsylvania, it is required that sulfuritic coal above the seam be segregated and covered with clean spoil. Similarly, cleaning plant reject can be dumped in the pit and covered in the operating procedures. At little extra cost, refuse can be carried on the empty haul from the cleaning plant to the pit.

Fig. 3 is an uncompacted edge seal which we have described in detail. It can be constructed cheaply in one operation with a shovel or dragline. The seal has been modified (as indicated) in accordance with observations on the erosion of clay seals; it is now recommended that each clay wall be set back 4 ft. (instead of 2 ft.) from the underlying wall, reducing the overall slope from about 68 to 52%.

Fig. 4 is a layer constructed, compacted edge seal, each wall of clay being 4 ft. high and set back from the underlying wall to provide an average face slope of 50%. It is a tighter seal and it offers more resistance to erosion than the preceding seal, particularly in clay-shale construction.

Although not indicated in the sketches, one would, if possible, construct the first wall by digging a trench and throwing the earth forward. The trench would be filled with the first layer of refuse. Clay for succeeding walls on a hillside pile would be obtained, as far as possible, from the hillside at the same level as the corresponding wall and excavated space would be filled with refuse.

In sealed layer construction, the seal for each layer should always be put down before the refuse. Otherwise, drivers invariably dump the refuse beyond its proper limit and the seal thickness is irregular and in some spots dangerously thin. Each layer should be set back from the underlying layer and the seals should be covered with vegetation to hold erosion to a minimum. (Liming may be necessary for the establishment of vegetation because of acid drainage.)

Fig. 5 is a partial edge seal suggested for economical use of a sealant of limited availability. Clay, fly ash, fine refuse or sludge from a settling pond may be used for the seal. The sealant is applied as a relatively thin layer between layers of refuse around the exposed edges of the pile, each layer extending inward from the edge for a distance of about 16 ft. In this manner, the sealant forms a series of dampers to check the draft along the slope; it is better protected from erosion than if it were applied as a thin external seal on the slope of the pile. In conjunction with slope reduction (to permit side compaction) this procedure should give satisfactory fire protection.

Crushed Refuse Piles

Some operators crush mine refuse and large size plant refuse to an intermediate size before disposal. This procedure is advisable particularly in the disposal of mine refuse in unsealed piles because large, unweathered rock and slate cannot be properly compacted.

Refuse from the cleaning plant has been finely crushed and deposited in layer constructed piles for many years at one Western Pennsylvania operation. Coal is crushed to −4 in. and is cleaned in a launder. The reject is passed over a shaker screen (3/8 in. square openings) and the overpass is crushed.

The feed to the screen is roughly as follows:

+3/8 in.	58%
3/8 x 20 mesh	35
−20 mesh	7

Crushed and uncrushed refuse go to the same bin and it is trucked from the bin to the disposal pile. Screen analysis of the refuse average as follows:

+3/8 in.	14.79%
3/8 x 1/4	24.51
1/4 x 20 mesh	47.47
−20 mesh	13.22

The refuse is deposited on the pile in separate heaps and it is leveled periodically with a bulldozer to a layer thickness of about 5 ft. The top of the pile is maintained very level, and it is compacted only by the trucks. Each layer is set back slightly from the underlying layer to reduce erosive effects. Rather deep drainage channels, however, are evident along the sides. Slight heating appears to have occurred in some fissures but there have never been any fires because all of the refuse is crushed to a fine size consist. The older piles are covered with crushed slag. Fire in these piles is extremely unlikely.

The cost of fine crushing is commonly considered to be excessive. However, the cost is justified in disposal operations where, in the absence of sealing clay and equipment for edge compaction, the best possible assurance is required that the refuse will not fire.

TREATMENT OF EXISTING REFUSE PILE FIRES

Water

Water, ancient aid to man in combating fire, appears to have been only partially successful in the extinguishment of refuse fires. During the Second World War, water was used extensively as a spray over burning tips in Great Britain to prevent their use as beacons for German airplanes. In most cases, the effect of the water was temporary; fires recurred when the spray was discontinued. A similar experience with attempted spray control of a fire in a long larry pile was noted near Pittsburgh. The fire appeared to be under control but it slowly spread toward the tipple. Finally, to halt its advance, a cut was made through the pile with a high pressure stream of water and a bulldozer. Advantageous use of the spray appears to be limited to situations demanding reduction of atmospheric pollution from piles inaccessible for effective leveling, grading, compacting and sealing operations (as in aerial tram piles). Similarly, water was observed in another instance to have been used with limited success by forming a basin on the top of a burning pile and filling it with water; complaints from neighboring residents subsided but the fire smoldered in cracks and fissures for many years. Better results can be obtained by pumping cleaning plant slurry into a basin on a burning pile. In the use of slurry, care should be taken that the water does not cut channels in the face of the pile.

Incidentally, water can be used very effectively in conjunction with a shovel

or bulldozer to quench, dig out or level and stamp out fires in accessible locations. In this manner, fires in a refuse area of about 10 acres were extinguished. Two pumps were installed, each capable of pumping 1000/gal./min. through 6 in. discharge lines. At the fire area, the 6 in. lines were split into a number of 2 in. lines in order to effect a wider distribution of water on the burning pile. As the fringe areas were cooled, the refuse was pushed aside with a bulldozer so that water could be applied directly to the burning areas. This procedure was followed until the entire area was extinguished in about 60 days.

Compacted Refuse Seal

A procedure for leveling the top, grading or doming the slopes, compacting and sealing the entire surface (including hot spots) with fresh refuse and extending the base where necessary to consolidate irregular portions of the pile has been followed. Intensive compaction procedures were used and control over the fires was gained without the use of clay. Needless to say, compaction of hot spots with a bulldozer and sheepsfoot roller must be attended with caution. In treacherous spots, the roller was pushed into the burning area ahead of the bulldozer. Generally, the slopes were graded and compacted but occasionally a procedure of envelopment was used, building a wall of fresh refuse around a burning slope and then filling in the hole; and some high slopes were not graded but were rolled along the top edge with the side roller previously mentioned, in this manner sealing fissures along the edge and forming a tight cap which greatly reduced the chimney effect and combustion along the slope.

Also, a refuse pile on the Allegheny River has been successfully reconstructed in a similar manner. Changing from larries to trucks for refuse disposal, the top of the old pile, very irregular and burning for many years, was leveled, graded, terraced and compacted and it was sealed with successive compacted layers of fresh refuse. At the same time, an extensive sealing operation was conducted over a period of about 2 years which resulted in a heavy clay seal over the face of the pile. This operation, completed in 1953, is another most creditable achievement in the extinguishment of gob fires.

Sludge Seal

Several refuse piles were extinguished by leveling the top and sealing the top and sides with cleaning plant sludge. The procedure was so successful that refuse is now dumped on a pile which was extinguished in that manner and they have continued to use sludge in the disposal operation to prevent the recurrence of fires. The sludge is trucked from the settling basin to the pile, and it is spread in a layer about 12 in. thick over each layer of refuse (about 5 ft. thick). The layer edges are also covered with sludge.

Fine Refuse, Fly Ash Seals

Refuse fires have been extinguished by sealing with – 1/4in. refuse. Other operators have obtained good results by sealing burning piles with cleaning plant sludge and with fly ash. It should be observed, however, that heavy seals of such fine substance are necessary because erosion at the normal angle of repose is quite severe.

Clay Seal

Generally, clay is of limited value when it is dumped over a burning area on the side of a pile because cracks develop in clay over hot spots and impair the seal. Clay can be used more effectively over surfaces which can be compacted. It may be noted, however, that at a pile now under observation a heavy crown of clay over the top and extending about half way down the steep slope appears to have confined a fire to its origin in the lower part of the pile.

OTHER CONSIDERATIONS

Trespassers

Trespass on the pile and particularly the activity of pickers should be prohibited. Treacherous hot spots and dangerous concentrations of carbon monoxides present special dangers to irresponsible persons. At surface level over fissures near hot spots concentrations of over 1000 ppm. of carbon monoxide (1500-2000 ppm. is a dangerous concentration for an exposure of 1 hr.) have been noticed. In no case, however, have appreciable concentrations of this gas been observed at a height of 6 ft. above the surface.

Vegetative Cover

Although the reclamation of stripped lands is receiving the attention of progressive operators, comparatively little interest has been shown in the establishment of vegetation on refuse piles. It will probably be found necessary to adapt the procedure to the character of the refuse. In general, efforts to promote vegetation are most likely to be successful (even with a thin cover of top soil) after a weathering period of such extent as may be necessary to leach the oxidation products of pyrite (sulphuric acid and ferric sulfate) and to establish moisture retention characteristics in some depth in the refuse.

Fresh Refuse on Old Piles

Many fires have resulted, it has been reported, from the piling of fresh refuse upon old refuse. This occurrence is most probable, it seems, when wet refuse is piled loosely against highly carbonaceous refuse, such as pickings from older operations. On the other hand, in reconstruction operations at several old burning piles, the piles have been leveled, graded and compacted (including hot spots) and then have been covered with layers of fresh, compacted refuse, and the fires have been smothered. Therefore, covering operations in which the refuse is compacted seem to be safe procedures but the placement of wet refuse beside old highly carbonaceous refuse may result in fires; and in the latter instance a barrier of clay should be interposed between the old and the new piles.

Mine Buildings

Buildings have been constructed on refuse piles at many old mines, particularly in valleys where the disposal problem is acute, where all available land near the mine has been covered with refuse and expanded operations has made it necessary to use refuse areas for buildings and storage of equipment and supplies. The disturbance of a dormant refuse pile occasioned by such construction may easily result in spontaneous heating and eventual firing of the old refuse pile. Such a fire may advance a considerable distance along exposed edges and into the uncompacted interior, even proceeding under a building with a concrete floor. Workers in the building may be subjected to dangerous concentrations of toxic gases and the foundation of the building may settle. It then becomes necessary to extinguish the fire and either repair the building or construct another building in some other location. Either procedure is troublesome and expensive. It is therefore desirable to avoid refuse areas for construction of buildings or, if construction is unavoidable, all exposed surfaces of the pile should be adequately sealed to prevent air entering the pile with resultant heating and firing of the interior.

Acknowledgment

The Committee gratefully acknowledges the assistance provided by the Report of Investigation by Dr. W. L. Nelson, Senior Fellow, Mellon Institute, from which this report was prepared.

Air Pollution Control Association
Technical Coordinating Committee
T-4—Coal

Richard C. Corey J. J. Foster
Richard B. Engdahl James R. Garvey
H. C. Rose, Chairman

General Report

Adopted by

National Conference on Metropolitan Problems

More than 250 persons including business men and women, labor leaders, public officials, educators, and officers in civic and professional organizations participated in the National Conference on Metropolitan Problems, held at Kellogg Center, Michigan State University, East Lansing, Michigan from April 29 to May 2, 1956. Executive Secretary H. C. Ballman represented APCA.

The full proceedings of the Conference are being prepared for publication.

At the National Conference on Metropolitan Problems, held at Kellogg Center, Michigan State University from April 29 to May 2, 1956, 200 business men and women, labor leaders, federal, state and local officials, educators, and officers of civic and professional organizations, divided into 12 separate groups and discussed for 3 half-day sessions (1) the metropolitan challenge to government;

(2) how to make government in metropolitan areas capable of doing its job, and

(3) what can be done to bring about greater cooperation among agencies, organizations, and individuals concerned with metropolitan problems.

The United States and Canada are rapidly becoming urban nations throughout this century, and especially since the war, most of the increase in population has occurred in the larger cities and their surburban fringes. Between 1950 and 1955, over 97% of the total increase of poulation in the United States was in the 168 standard metropolitan areas listed in 1950 census.

For purposes of conference discussion, the standard metropolitan area, as defined by the federal government[1], was generally accepted as satisfactory for the gathering, summation, and comparison of statistics. It was felt, however, that uncritical use of this definition would hide many essential elements in the metropolitan complex. To one concerned with the government of metropolitan areas essential elements are: large size, high population density, interdependence, fluidity of movement, and fractionated government.

There are many urban problems that are not metropolitan problems. There was general agreement to consider as *metropolitan problems* only those problems arising from a large congested population living and working interdependently in a considerable territory and rushing to and fro, with governments which do not coincide with the patterns of life.

Small urban places are, however, developing many of the political and governmental characteristics of the large metropolis.

Development of metropolitan communities is the natural end-product of over a century of industrialization accompanied by increased agricultural productivity. The form of the metropolitan area is determined by such factors as changing technology (the automobile, the septic tank, the power pump, *etc.*) economics of mass housing, lack of room in the central city and older suburbs, government housing policies, search for individual homes surrounded by acreage, *etc.* There was considerable speculation as to whether the future metropolitan community would look like New York or Los Angeles.

The almost-universal division of metropolitan areas into many units of local government is the result of numerous factors. In general, it reflects the fact that population growth has extended outward from urban centers and overrun boundaries of the central city and many other long-established governmental entities. In some metropolitan areas, cities originally separated by open country have simply grown into each other. Also, additional local governmental agencies have been created in growth areas to provide urban services as needs become critical.

Rivalries and conflicts among neighboring communities, suburban distrust of the central city, as well as general lack of foresight, have contributed to the absence or failure of measures which might have limited or relieved effects of metropolitan fragmentation.

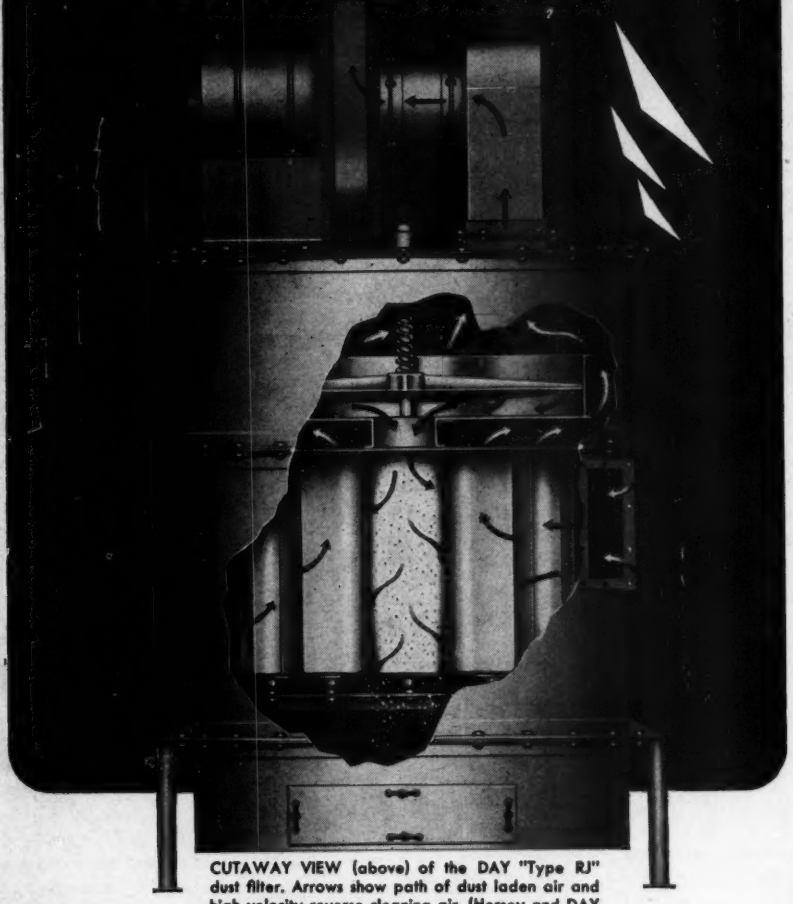
The growth of existing metropolitan areas and development of additional areas that meet the definition used by the Bureau of the Census will continue for several decades. By 1960 there may be a score of new standard metropolitan areas. Present and new metropolitan areas will account for most of the increase in population. This enormous growth will increase the number and complexity of metropolitan problems.

This growth will be accompanied by further concentration of the labor force, higher and more widely distributed income, more leisure time, larger families more old people, and more demand for the services of local governments. Development of air travel may affect distribution of population as will continued movement of surplus farm population to cities.

Development and expansion will continue to take place on fringes of metropolitan areas. One result in some parts of the country will be the coalescence of metropolitan areas into vast urban regions. Some participants pointed out that the *linear city* might invalidate in

[1] Except in New England, a standard metropolitan area is a county or group of contiguous counties which contains at least 1 city of 50,000 inhabitants or more. In addition to the county, or counties, containing such a city, or cities, contiguous counties are included in a standard metropolitan area if, according to certain criteria they are essentially metropolitan in character and socially and economically integrated with the central city . . . In New England . . . towns and cities *instead of counties* were the units used in defining standard metropolitan areas—*U. S. Census of Population* (1950) Vol. I. *Number of Inhabitants. p.* 33.

NEW
DUST FILTER DEVELOPMENT



CUTAWAY VIEW (above) of the DAY "Type RJ" dust filter. Arrows show path of dust laden air and high velocity reverse cleaning air. (Hersey and DAY patents applied for.)

DAY "Type RJ" Dust Filter—First and Only Dust Filter to Offer All These Advantages

- An economical, high efficiency, automatic-continuous dust filter.
- Incorporates Hersey principle of high velocity reverse air for filter sleeve cleaning in a low cost unit.
- Provides efficiencies as high as 99.99+%.
- Automatic—continuous operation makes filter ideal for controlling dust created by continuous process equipment.
- Economical operation—utilizes ½ HP motor for filter sleeve cleaning.
- High air handling capacity for small space occupied by filter.
- Can be furnished for pressure or vacuum operation.
- Shipped completely assembled. Only piping and electrical connections required for operation.

The DAY "Type RJ" dust filter has many advantages to offer your plant. Get the complete facts, write toDAY for Bulletin 560.

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some areas the concept of central city plus surburbs; we may be in danger of attacking a problem already outmoded. However, it is noted that a few states and the federal government are beginning to deal with some urban problems on regional and national scales.

The function of the core city, or at least the central business district, is changing. What these changes will mean a few years from now is not clear; it appears that even with a new role to play in the social, economic, and political life of the metropolis the core city will continue as a center of metropolitan life. Physical and functional obsolescence of older areas may continue at an accelerating rate unless checked by urban redevelopment and renewal.

No major counter-trends can be identified, although traffic congestion and the possibility of aerial warfare could in time reverse the movement of poulation and industry to metropolitan areas.

Conference participants recognize problems vary in extent and intensity in different metropolitan areas and occur at different times in the same metropolitan area. Some participants consider the diffusion of popular control of metropolitan activities the most important metropolitan problem. Others while admitting the importance of this problem, emphasize the primary importance of providing a sufficient level of service to urban people. Transportation, water supply and distribution, disposal of sewage and other wastes, schools, land use planning and control are pressing problems in most metropolitan areas.

Failure to solve these problems affects urban life adversely at many points. Economic productivity of metropolitan areas is reduced and their full potential development is unrealized. There is inequitable financing of public service. The comfort and convenience of the public is reduced. The ineffectiveness of local self-government increases the drive toward direct state and federal action.

It is not possible at this point to chart fully the course or courses that will lead to the best solutions. There are, however, certain clear objectives which will guide us in determining governmental structures and strategy needed in metropolitan areas.

The basic need is for government that will serve the people of metropolitan communities effectively and efficiently with active citizen participation and popular control. An adequate and equitable revenue system, some local initiative and self-government for traditional or natural communities within a metropolitan area, and provision for adaptation to growth and change are important additional objectives. Any system of metro-

politan government, moreover, should be designed and operated with a view to the sound economic and social development of the area as a whole, and the full utilization of private initiative and participation in meeting community needs.

There is no single form of governmental organization that will meet the needs of all metropolitan communities. Fortunately, however, we have had experience with a variety of arrangements which, in different combinations, offer hopeful avenues to progress.

Government must be tailored to the needs of each community. This means local people and officials must assume major responsibility for studying their own needs and forming and executing their own plans. The possible scope of local action depends to a very large extent on the state.

There is an urgent need for review and relaxation of certain constitutional and other legal limitations on the powers of existing local units and for enactment of legislation to permit new forms of local government. Development of local capacity to finance government in metropolitan areas adequately and equitably calls in many places for elimination or modification of legal limitations on fiscal powers as well as for revision of local tax systems, including assessment practices.

While complete consolidation of all local governments into a new metropolitan government is one of the theoretical possibilities, it is neither a likely nor a desirable solution, except possibly in new and relatively small or emerging metropolitan communities. Serious consideration should, however, be given to development of general metropolitan governments with limited functions, leaving all other functions to existing local units. This goal may be approached in different ways in different communities. Greater use is being made, for example, of the urban county as an instrumentality of metropolitan government, especially where a single county embraces all or most of the metropolitan area. Single or multi-purpose metropolitan districts and authorities are demonstrably useful in meeting urgent service needs. Yet establishment of a variety of such agencies in an area increases the number of units of government, complicates the problem of achieving coordinated action, and increases the difficulty of maintaining effective popular control. It is possible, however, that development of a multi-purpose metropolitan district to meet the immediate and urgent need for area-wide services might lead ultimately to establishment of a general government carrying on a limited number of metropolitan functions.

Ducon Centrifugal Air Washer TYPE WB

. . . a Ducon engineered recovery system saves thousands of dollars in wallboard production.

A leading manufacturer of wallboard experienced this problem, losing valuable profits in waste materials which escaped not only in hole drilling but during sizing, trimming, beveling and mortising operations. To combat this problem Ducon engineers recommended a special installation which provided efficient, automatic collection of waste materials and effected their return to the system for reprocessing.

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The responsibility of the state does not stop with the relaxation of legal limitations or enactment of facilitating legislation. The state has an obligation to provide active leadership in the solution of metropolitan problems. This might begin with establishment of a temporary state commission to determine the need for and the nature of appropriate state action. States already provide various kinds of guidance and technical assistance to local authorities. States should consider expansion of such activities, including continuous research and information concerning metropolitan problems and intergovernmental relationships. For this purpose, the establishment of a state department of local government or urban affairs is worthy of consideration.

One of the most important needs in any metropolitan area is some form of over-all metropolitan planning. The method of organizing and the extent of such planning must be fitted to each area, but the state can play an important role by means of technical assistance.

While the state has primary responsibility for local, and consequently for metropolitan government, the federal government also has a role in the solution of metropolitan problems. More effective coordination of the presently dispersed activities of the federal government within and affecting metropolitan areas would make it easier for state and local governments to deal with metropolitan problems in area-wide terms.

A sense of community is essential to success of metropolitan government, however organized. Citizens and officials must learn to think and act in terms of their responsibility to the whole area as well as to any smaller community within it to which they may belong.

This need places a great responsibility on all organizations and groups, official or unofficial, interested in metropolitan government. It calls upon them to sponsor and promote a many-sided program of research, education and action, designed to clarify the essential nature of the relationships of the individual to the communities in which he lives and moves, and implications of these relationships for government.

Many suggestions for basic and operational research were made and recorded. These suggestions will be analyzed and reported to members of the conference and interested researchers.

Participants in this conference feel their coming together has substantially enhanced their awareness of metropolitan problems and their sense of membership in their metropolitan communities. They intend to continue and expand this association. They are convinced that metropolitan areas are generating the most important problems of domestic government facing the country today. These problems challenge our faith in basic democratic principles and test our ability to adapt political institutions and practices to the rapidly changing requirements of modern society. Our handling of them will vitally affect the livability of our homes, the health and efficiency of our communities, the productivity and prosperity of our economy, the soundness of our national defense and the general welfare and happiness of our people. The search for solutions is urgent

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Recommendations of Subcommittee Concerning Further Activities

This subcommittee, appointed to consider and offer recommendations to this Conference concerning its further activities, submits its report as follows:

(1) The work done prior to this conference, the discussions that have taken place here, and the conclusions arrived at by those in attendance, indicate a very real need for a greater degree of cooperation among the organizations, agencies, and individuals concerned with the pressing problems which face the metropolitan areas of our country.

(2) The solution of those problems by the development of such cooperation would be greatly advanced by the establishment of a Continuing National Conference on Metropolitan Problems.

(3) Membership in or affiliation with the conference should be available to organizations and agencies interested and active in metropolitan problems; consideration should be given to the creation of a form of membership for qualified individuals on a basis that will be in accord with the purposes of the conference.

(4) The primary functions of the conference should be to serve as a cooperating agency for groups and organizations concerned with metropolitan problems; to encourage and cooperate in such research, and to prepare and cooperate in the preparation of such publications and to hold such national, regional and other meetings as may further the objectives of the Conference. All of this should be done without duplication of effort and by the use of existing agencies, in so far as possible.

Therefore, this subcommittee recommends that a committee for the aforesaid purpose be chosen in such manner as this conference may determine and that Frank Moore, who deserves so much credit for the success of this conference, be requested to serve as chairman of this committee and of the continuing conference.

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DUST AND FUME CONTROL
INDUSTRIAL VENTILATION
STACK DUST EMISSION MEASUREMENTS
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AIR ANALYSIS

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AIR POLLUTION...
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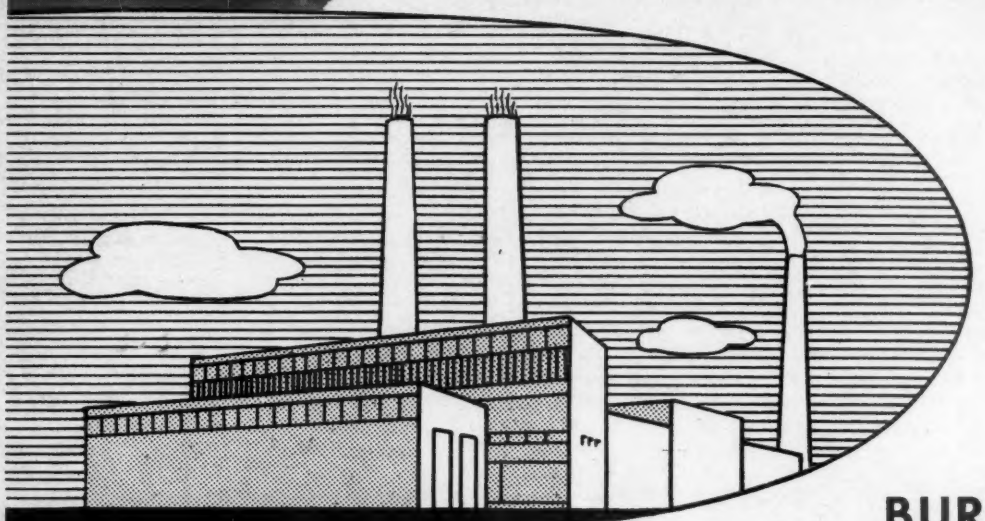

BURNING...
IS NOT NECESSARILY INCINERATION

What is PROPER incineration? It's the complete destruction without accompanying smoke, fumes, odors, fly ash and other annoying factors which can put burning into the nuisance class. Burning, in itself, is not necessarily incineration. Just a furnace won't do.

When and where, however, does the design of the incinerator start? Not until answers are found to many questions involving such important factors as:

a . . . Are the wastes amenable to incineration? Are they dry? Wet? Contaminated? Volatile? Do they have high heat content? Are they pumpable?

b . . . Location of incinerator? Air supply?

c . . . How will wastes be handled? How charged?

d . . . Operating schedule?

e . . . Will surroundings dictate minimum height of stack?

f . . . Local codes and ordinances?

Who knows best what factors to ferret out and what answers need to be sought? Who knows best how to integrate various influencing factors. Our design and construction business for more than sixty-five years has been in this very field of waste-disposal through proper incineration. We're known as Incineration Specialists. Our long experience is available to you, to your engineering consultant and to your architect for the asking WITHOUT OBLIGATION. We have competent representatives in 85 cities in United States and Canada. We'll be glad to have the one nearest you call and help you get started on your problem.

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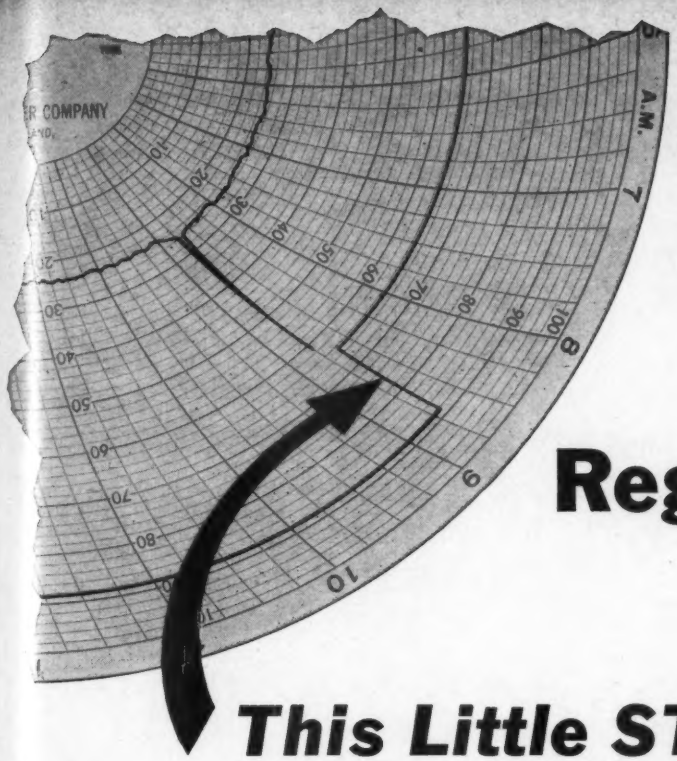

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This record, combined On The Same 24-Hour Chart with a continuous measurement of Smoke Density, gives *a complete permanent record of performance*—visual proof, in an unbeatable pair, of your efforts to comply with the Air Pollution Control Ordinances.

Now you can have such a record—and at a very moderate cost—with the new *Bailey Running Time Recorder* combined with the Bailey Smoke Density Recorder.

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The industry's most experienced organization in dust, fume and fly ash recovery

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Because the Western Precipitation Corporation stands alone in its years of continuous leadership in the complex science of recovering suspensions from industrial gases, Western Precipitation installations, quite naturally, are recognized throughout the world as the best obtainable. . .

■ Almost a half-century ago—in 1907 to be exact—Western Precipitation installed the first commercial application of the now-famous Cottrell Electrical Precipitator—and has more know-how, more varied experience and application background in world-wide installations in this type of equipment than any other organization, domestic or foreign.

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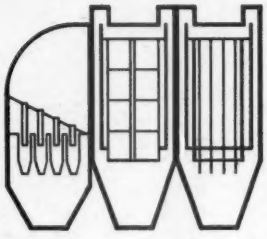

■ Western Precipitation was the first to combine Electrical and Mechanical recovery advantages in one compact, coordinated system—the CMP (Combination Multiclone—Precipitator) Unit. This equipment, offering almost constant collection efficiency despite varying gas volume, requires years of experience in both electrical and mechanical recovery methods for proper operating "balance".

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For sampling city atmosphere for determination of smoke and haze concentration, the A.I.S.I. Smoke Sampler is recommended. It may be employed for two purposes:

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The low cost of the A.I.S.I. Automatic Smoke Sampler makes it feasible to locate a large number of them for complete coverage of a geographical area, and its quietness overcomes any objections to its location in or near individual residences.

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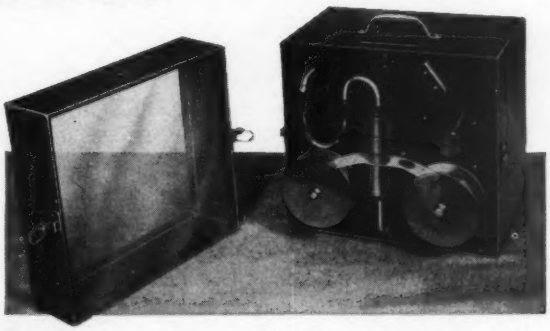

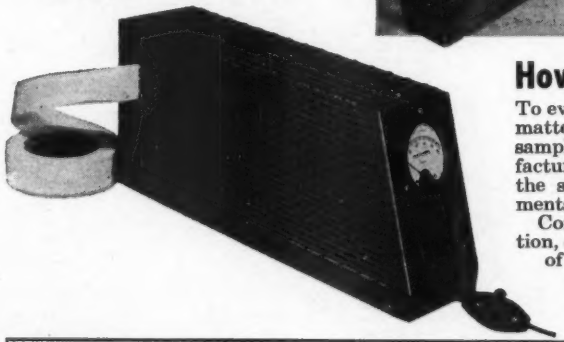

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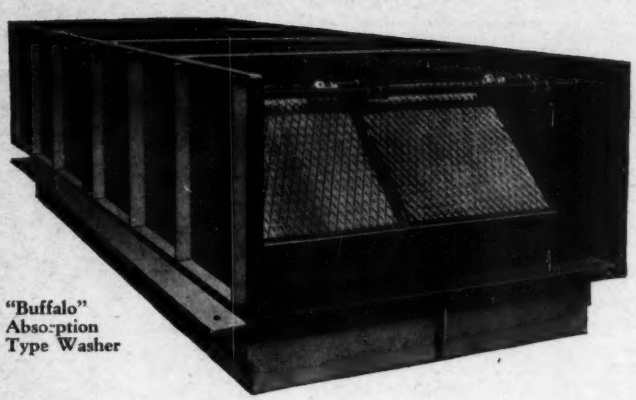

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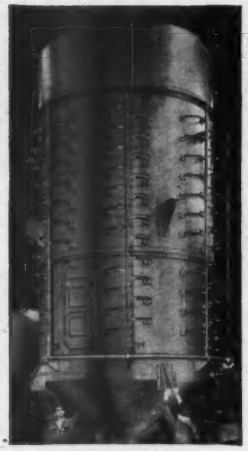
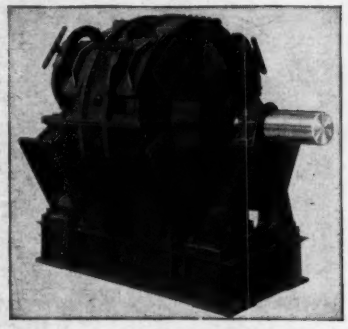


"Buffalo" Hydraulic Scrubbing Tower



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"Buffalo" equipment includes Air Washers, Hydraulic Scrubbing Towers, Hydro-Volute Scrubbers, Rotary Multi-stage Gas Scrubbers, Gas Absorbers, Blowers, Exhausters and Pumps to solve most any cleaning problem you may have — all engineered to the job, for highest efficiency and simplest possible maintenance. Call on the "Buffalo" Air Engineer in any principal city for sound, helpful recommendations.

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Abstract from Paper 56-13, "Air Pollution Control by Activated Carbon" Annual Meeting of Air Pollution Control Association, Buffalo, N. Y., May 20-24, 1956. Delivered by H. L. Barnebey, Vice President, the Barnebey-Cheney Company, Columbus, Ohio.

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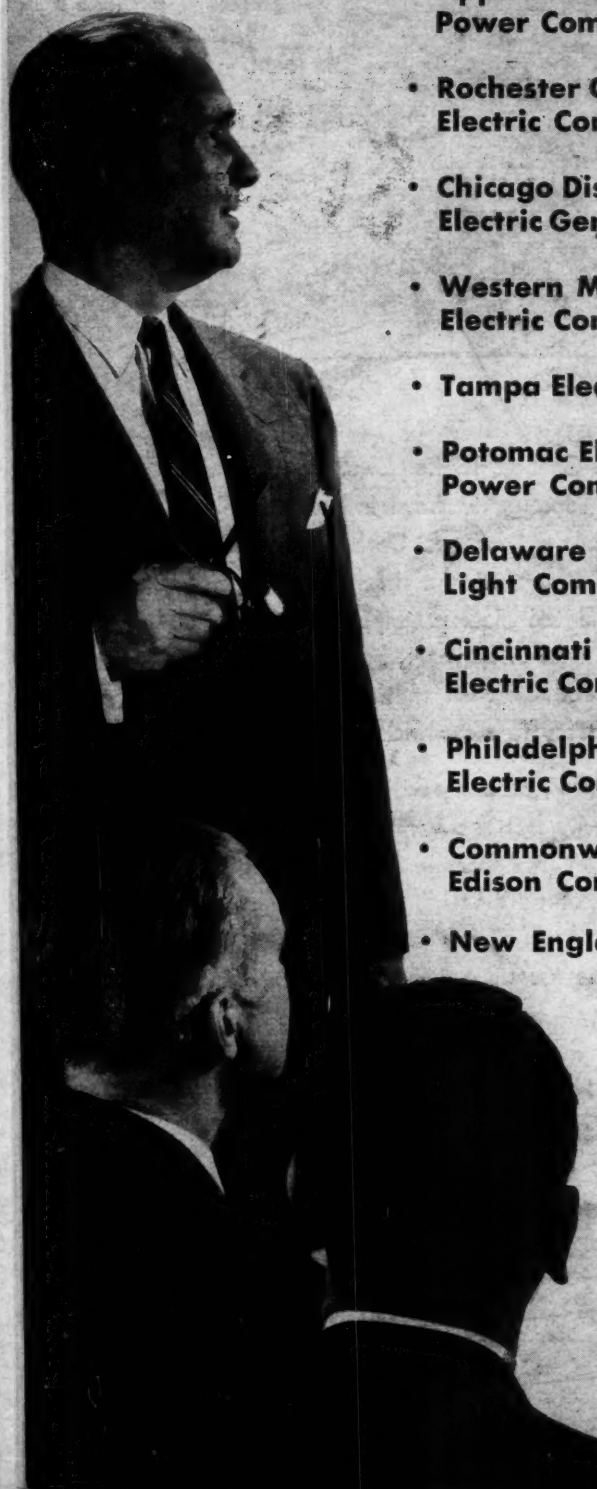

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• Western Massachusetts Electric Company	West Springfield	3
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• Delaware Power & Light Company	Indian River	1
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